

FERROMAGNETIC RESONANCE AND RELAXATION

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for the Degree of
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**BY
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TO
MY MOTHER
WHO IS NO MORE

Certified that the work presented in this thesis
has been carried out by Pooran Singh under my supervision

A handwritten signature in dark ink, appearing to read 'J. Mahanty', with a long horizontal stroke extending to the left.

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SYNOPSIS

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In the work presented here different models employed to describe ferromagnetic resonance (FMR) and ferromagnetic relaxation (hereafter referred to as relaxation) have been briefly discussed. The "s-d exchange" interaction model between ferromagnetic 3d electrons and conduction s electrons in a ferromagnetic transition metal (Ni, Fe) (hereafter referred to as ferromagnetic metal) has been studied in detail to explain relaxation in these metals. The coupling of spin waves (magnons) with lattice vibrations (phonons) resulting from magnetostriction has been considered and a two magnon - one phonon process has been studied. The renormalized spin wave and phonon energies have been obtained. The method of double-time temperature dependent Green functions has been used to investigate these effects

Chapter I gives a brief account of the present state of our knowledge of FMR and relaxation, both in ferromagnetic insulators and metals. FMR and relaxation in ferromagnetic insulators is well understood in terms of various magnon-magnon and magnon-phonon processes. However, the linewidths and relaxation times in ferromagnetic metals are not explained on the basis of these processes. In a ferromagnetic metal the linewidth is believed to be due to (1) magnetic relaxation damping - the microscopic origin of which is not exactly known. This is described macroscopically by including Landau-Lifshitz or Bloch-Bloembergen type of damping terms in the equation of motion for the magnetization of the system. (11) exchange conductivity broadening - which arises because of the fact that the exchange stiffness opposes the tendency of the dynamic magnetization to vary within a small skin depth. The linewidths and relaxation times in ferromagnetic metals are not explained on the basis of these two mechanisms even

In Chapter II Kittel's phenomenological theory of FMR has been briefly discussed. The expression for the FMR frequency has also been derived quantum mechanically as obtained by Van Vleck. The effect of the shape of the specimen, magnetic anisotropy and domain structure on the FMR frequency has been qualitatively discussed. The phenomenological approach to damping has also been briefly described. The phenomenological theory satisfactorily explains the shape of the resonance curve but is inadequate

to explain linewidths and their temperature dependence. It is not surprising as the microscopic nature of the relaxation mechanism is not exactly known.

Chapter III presents a brief introduction to the Green function formalism that has been used in this and the subsequent chapter. The method is applied to the Heisenberg model of a ferromagnet with demagnetizing energy as first considered by Tyablikov. It is noticed that in the approximation (random phase approximation) considered here the Green functions have poles on the real axis and consequently there is no damping. The FMR frequency formula obtained is applicable at all temperatures.

The coupling of the magnons with phonons has been considered and a two magnon-one phonon process has been studied. The renormalized magnon and phonon energies are obtained.

In Chapter IV the spin waves in an itinerant electron model have been discussed. The interaction of the ferromagnetic 3d electrons interacting with conduction s electrons by means of "s-d exchange" interaction has been considered. The expressions for the dynamic magnetic susceptibility and for the linewidth due to "s-d exchange" interaction have been obtained. It has been suggested that a bunch of magnons participate in the relaxation process resulting in the conduction electron spin flip.

In the concluding Chapter V, the results of the present investigation are summarized. The limitations of the theory are also discussed.

CHAPTER - I

INTRODUCTION

Ferromagnetic resonance (FMR) is a powerful experimental technique for studying the modes of excitations of a ferromagnet and the lifetimes of modes arising out of their coupling with other excitations. The basic principle of the experiment is as follows

A ferromagnet is placed in a static magnetic field and an oscillating radiation field is applied perpendicularly to the constant field*. As the frequency of the radiation field is varied, resonance occurs due to absorption of energy from the radiation field at a particular frequency depending on the value of the constant field. The ferromagnetic resonance of this type was first reported by Griffiths¹ in 1946. It differs from other kinds of magnetic resonance such as electron paramagnetic resonance in that here the collective effects are involved. All the spins are parallel and collectively precess about the static field in phase. It is also called uniform-mode ferromagnetic resonance. In the framework of the spin wave theory the uniform precessional mode corresponds to excitation of spin waves of wave vector $k = 0$. (The excitation of spin waves of $k \neq 0$ requires rather special conditions. These non-uniform ($k \neq 0$) modes are excited in spin wave

*There is a different type of experiment in which r.f. field is applied parallel to the static field. This is called a parallel pumping experiment.

resonance experiments. These experiments were suggested by Kittel and first observed by Seavey and Tannenwald²).

In the present work we would restrict our discussion to the uniform mode ferromagnetic resonance. The occurrence of this resonance is due to the transition between the state of the system with total spin NS (where N is the number of lattice points and S is the atomic spin) aligned along the magnetic field direction and the state with all the spins reversed, i.e., with total spin $-NS$. The transition is caused by the exciting radiation field.

The first experiments on FMR done on ferromagnetic metal (Ni, Fe, Co) thin films by Griffiths¹ indicated that the absorption curve had a resonance behaviour with a maximum for a certain value of the magnetic field which depended on the frequency. The interpretation which he gave suggested that the resonant absorption by the magnetic dipoles (which might, of course, be multiples of μ_B , where μ_B is the Bohr magneton) was taking place leading to a loss of energy by the radiation field. The resonant frequencies, he observed, were greater than the calculated Larmor frequencies for electron spin by factors of about two to six. Griffiths unsuccessfully tried to explain this anomaly by introducing Lorentz field, an approach which was not justified. A correct theory was later given by Kittel³ in 1947 which led to the values of the resonant frequencies in close agreement with experimental determination. Kittel showed that it was

important to consider the dynamic coupling caused by the demagnetization field normal to the surface of the specimen. The principal result of his theory was that the appropriate Larmor frequencies should be calculated as for a field strength $(BB_1)^{\frac{1}{2}}$ rather than for B , the static magnetic field, where B_1 is the magnetic induction

The width of the resonance line arises out of the interaction of the uniform mode with other modes (such as magnons, phonons and electrons) in the system. The origin of the width of the ferromagnetic resonance lines is not clearly understood specially in metals

We give below a brief account of some experiments on FMR which mainly deal with the measurements on linewidths and relaxation times and their interpretation both in insulators and metals

Insulators

In non-conducting ferrimagnetic single crystals FMR ($k \approx 0$) linewidth can be very narrow. LeCraw and Spencer⁴ who measured linewidths in pure yttrium iron garnets (YIG) found that the linewidth can be as small as 0.2 oersteds at 3.3 KMC/sec

The dominant relaxation mechanism in these materials appears to be magnon-magnon interactions. For a detailed discussion of the various multiple magnon processes we refer to Haas and Callen⁵ and to Sparks⁶.

In any resonance experiment one might expect some broadening of the resonance line due to specimen inhomogeneities or surface imperfections. LeCraw et al.⁷ showed that the ferromagnetic resonance linewidths of yttrium iron garnet spheres depended on the size of the grit used for polishing the specimens and hence to the surface inhomogeneities. Sparks et al.⁸ have proposed that the two magnon scattering of $k \neq 0$ magnons plays an important role in the relaxation of imperfectly polished YIG.

Spencer and LeCraw⁹ have measured the longitudinal relaxation time τ_1 in YIG as a function of temperature. They find that at 9.34 KMC/s between 2° and 30°K

$$\frac{1}{\tau_1} \approx 10^5 \text{ T sec}^{-1}$$

which is in good agreement with the value calculated by Sparks and Kittel¹⁰

Various types of volume inhomogeneities can also affect the line width¹¹.

In principle, the magnon-phonon interactions should also contribute to the line width and a direct process would occur when the phonon and magnon frequencies and wavelengths are matched. In the "cross-over" region (where magnon and phonon frequencies and wavelengths are equal) the magnons experience large magnon-phonon interaction and this results in a marked increase in the ΔH_k of these magnons where ΔH_k

is the width of the spin wave resonance. An increase in ΔH_k will be reflected in an increase in the critical r.f. field strength necessary to initiate unstable growth of these spin waves. Turner¹² has found that in YIG the critical r.f. field in a parallel pumping experiment ($B \parallel B_x$) does indeed increase sharply for certain values of the d.c. field, that is, for certain values of k . Akhiezer¹³ and, Kittel and Abrahams¹⁴ have theoretically investigated phonon-magnon processes in detail.

Yttrium iron garnets containing rare earth impurities have also been experimentally studied. The FMR linewidths of rare earth garnets and of YIG containing rare earth impurities differ markedly^{15,16} from those of the pure YIG in that the linewidths of the rare earth garnets are very large. In addition, the linewidth has a very strong temperature dependence. The linewidths have been explained on the basis of magnon-phonon scattering induced by rapidly and slowly relaxing rare earth ions^{17,18}.

Relaxation processes involving transfer of valence electrons between ions of variable valency such as ferric and ferrous ions have also been studied in garnets.⁵

One can fairly claim that the linewidths in ferromagnetic insulators and ferrites are well understood in terms of magnon-magnon and magnon-phonon processes, which are induced by dipolar interactions and magnetostriction.

In the case of metals, however, the situation is not very clear. The electromagnetic waves enter a narrow surface layer of the metal called the skin depth and consequently the direction of magnetization vector \vec{M} is not uniform and the effective exchange field is not parallel to \vec{M} and a torque acts on \vec{M} which modifies the equation of motion of \vec{M} and the nature of the resonance. This modified resonance is called "spin wave resonance" following Rado and Weertman¹⁹. As stated earlier, these spin wave experiments were first suggested by Kittel²⁰ and were first observed by Seavey and Tannenwald²

In very carefully prepared specimens of Co, Ni and Fe, the narrowest spin wave linewidths that have been observed by Rodbell²¹, Phillips and Rosenberg²² are of the order of 30 - 50 oersteds.

The linewidth measurements on perfect metal single crystals have been made by several other authors^{21,23,24}. The relaxation times obtained from their measurements cannot be explained on the basis of spin-spin^{25,26} and direct spin lattice relaxation^{13,27,28}. They, however, show that the relaxation of the spin waves excited at FMR contributes to the linewidth in addition to the exchange broadening caused by inhomogeneity of magnetization in the skin depth²⁹. Even if surface pinning of spins is included, this result remains unaffected³⁰.

More recent and detailed measurements of linewidths on pure iron are by Bhagat et al^{31,32}. The widths in iron

are found³¹ to vary as the square root of the frequency and are due mainly to exchange conductivity mechanism. They were also led to conclude on the basis of their observations that for iron the relaxation frequency λ is essentially temperature independent from 4°K to 1000°K

In the case of Ni, more recently linewidth measurements have been reported by Rodbell³³, Salamon³⁴, and Bhagat et al^{32,35}. The linewidths in Ni are enormously large (of the order of a few thousand oersteds) The results obtained by these authors are not all in agreement The widths for Ni can be accounted for by relaxation damping described by a frequency independent Landau-Lifshitz coefficient λ ($= 2.3 \cdot 10^8 \text{ sec}^{-1}$) plus a smaller contribution from the exchange conductivity mechanism³¹ Salamon³⁴ finds an effective λ (relaxation frequency) decreasing with increasing temperature. This result is in qualitative agreement with Bhagat et al³⁵. Salamon also finds an increase in g value as he goes from below T_c to above T_c . In the case of Bhagat et al, the increase in g is rather gradual. The reasons for this discrepancy are not clear.

In order to understand the mechanism of FMR, it is necessary to consider various models that have been used to describe it The important models are the following

Spin Wave Model

This model assumes that the system is very near to its ground state. It is, therefore, particularly valid at

low temperatures when the departure of the ferrimagnet from its ground state (when all its spins are aligned parallel to each other) is very slight. Van Kranendonk et al.³⁶ used this model to obtain the frequency of the uniform ferromagnetic resonance. The explicit structure³⁶ of the Hamiltonian used is the following

$$H = g\mu_B B \sum_i S_i^z - 2J \sum_{i>j} \vec{S}_i \cdot \vec{S}_j + \sum_{i>j} D_{ij} \left[\vec{S}_i \cdot \vec{S}_j - \frac{3(\vec{S}_i \cdot \vec{r}_{ij})(\vec{S}_j \cdot \vec{r}_{ij})}{r_{ij}^2} \right] \quad (1)$$

where $D_{ij} = \frac{g^2 \mu_B^2}{r_{ij}^3}$, r_{ij} denotes the vector joining the atoms i and j , μ_B is the Bohr magneton, B is the constant magnetic field, J denotes the isotropic exchange integral, S_i denotes the atomic spin at the i^{th} lattice site. The various terms in equation (1) represent respectively the Zeeman energy, Heisenberg's exchange energy and the dipolar energy.

The interaction of the radiation field with the spin system assuming that the radiation field B_x is uniform throughout the sample (i.e., the skin depth is longer compared to the dimensions of the specimen under investigation) has the form given by

$$H_{\text{int}}^t = g\mu_B B_x \sum_i S_i^x \quad (2)$$

where the sum is over all the lattice sites of the crystal. J.T

The radiation field thus couples only with the transverse components of the magnetization

The interaction (2) causes transitions between different energy levels of the ferromagnetic electron states and excites spin waves of wave vector $k = 0$ (called uniform precession mode) as can be seen by replacing S_i^x by $S^{\frac{1}{2}}q_1$ in the harmonic oscillator approximation, q_1 being the coordinate operator. Thus

$$H'_{int} = g \mu_B B S^2 \sum_1 q_1 = g \mu_B B (NS)^{\frac{1}{2}} q_0 \quad (3)$$

where $q_0 = \frac{1}{N} \sum_1 q_1$, is the completely symmetrical normal coordinate. Thus the radiation field excites only the uniform precessional mode.

Van Kranendonk et al³⁶ using harmonic oscillator approximation obtained the normal modes of the system. The spin waves which are the elementary excitations are the normal modes of the spin system. The energy eigenstates of the system are characterized by n_k , the number of excited spin waves with wave number k . Since the $k = 0$ spin waves are excited at FMR, the frequency of this mode is given by

$$\omega_0 = g \mu_B \left[\left\{ B + (N_x - N_z) \mu_s \right\} \left\{ B + (N_y - N_z) \mu_s \right\} \right]^{\frac{1}{2}} \quad (4)$$

This result is identical with Kittel's expression except that the magnetization M is replaced by the saturation magnetization value M_s . This is expected as the spin wave model gives the low temperature value of the FMR frequency.

The model Hamiltonian equation (1) is based on the assumption that the electrons are localized on the atoms at the lattice sites. It is particularly suited to the case of ferromagnetic insulators where the localized spin picture is applicable. The localized spin model may also be used to treat rare earth metals because in them the 4f shell is well within the atom and the electrons can be regarded as localized. The situation in ferromagnetic transition metals is different. Here the 3d shell is on the outside of the atom and the 3d electron wavefunctions of the neighbouring ions overlap and thus form a narrow band of states. This means that the 3d electrons move from one atom to the other and, therefore, the localized electron concept is not applicable. This is called an itinerant (band) electron model³⁷⁻³⁹. This is based on the competition between the kinetic energy of the electrons in a band and the electron exchange in the Hartree-Fock approximation. This exchange may be thought of as arising from the intra-atomic rather than inter-atomic exchange. Rajagopal et al⁴⁰ have studied the magnetic properties of itinerant electrons in detail. We shall discuss spin wave excitations in this model in Chapter IV.

Various modifications of these itinerant and localized models have also been used to treat real solids. One such model is the so called "s-d exchange" model. This has been used to investigate linewidths and relaxation times in

ferromagnetic metals It was first considered by Zener⁴¹ Kasuya⁴² showed that the exchange interaction between the 3d electrons is mediated by s electrons and leads to an effective Hamiltonian for d electrons The Hamiltonian for the magnetic 3d spins exchanged coupled to s band electrons can be written in the form^{43,44}

$$H = H_0 + H_{s-d}$$

$$H_0 = \sum_q \epsilon_q c_q^\dagger c_q - \sum_{m \neq n} J(\vec{m} - \vec{n}) \vec{S}_m \cdot \vec{S}_n + 2 \mu_B B \sum_n S_n^z$$

$$\text{and } H_{s-d} = -2 \sum_{mn} J_{s-d}(\vec{r}_m - \vec{r}_n) s_m \vec{S}_n$$

$$\text{where } \epsilon_q = \epsilon_q^0 - s J_{s-d}(qq) + 2 \sigma \mu_B B \quad (5)$$

$$\epsilon_q^0 = \frac{\hbar^2 q^2}{2m} \quad \text{is the renormalized s electron energy}$$

c_q^\dagger and c_q denote creation and annihilation operators for s-band electrons. $J(\vec{m} - \vec{n})$ and J_{s-d} denote the d-d and s-d exchange integrals. In this model also the 3d electrons have been assumed to be localized which is not justified as has been pointed out earlier.

By writing the Hamiltonian (5) in second quantized form and also using spin wave approximation, it can be shown^{43,44} that the "s-d exchange" interaction brings about the coupling of the spin waves and conduction electrons which are the elementary excitations of the solid in this model The spin waves can relax through their interactions with conduction electrons if they (spin waves) have sufficient energy (of the

order of J_{s-d} required to flip the conduction electron spin) This model has been examined in Chapter IV of the present work in detail.

The spin waves can also couple with lattice vibrations (phonons). This magnon-phonon coupling can be brought about by the magnetic dipolar interaction⁴⁵ and/or magnetostriction⁴⁶. A two magnon - one phonon process has been studied in Chapter III.

Phenomenological Model

We know that the equation of motion of the magnetization \vec{M} can be written as follows

$$\frac{d\vec{M}}{dt} = \gamma [\vec{M} \times \vec{B}] \quad (6)$$

where $\gamma = \frac{ge}{2mc}$ is the spectroscopic splitting factor. This is the undamped equation of motion for \vec{M} . To explain resonance linewidths it is very essential to include damping into the equation of motion. The first phenomenological equation was suggested by Landau and Lifshitz⁴⁷ and Gilbert⁴⁸ and reads as follows⁴⁹

$$\begin{aligned} \frac{d\vec{M}}{dt} = & \gamma [\vec{M} \times \vec{B}] + \gamma [\text{Anisotropy torque}] + \frac{2A\gamma}{M_s} \vec{M} \times \nabla^2 \vec{M} \\ & - \frac{\lambda}{\gamma M_s^2} \vec{M} \times \frac{d}{dt} \vec{M} \end{aligned} \quad (7)$$

where M_s and A are respectively saturation magnetization and exchange stiffness. λ is the relaxation parameter. The terms in (7) respectively denote the torque due to external

(static) field, anisotropy torque, the exchange field torque and the spin lattice torque. By working with a macroscopic dynamical equation one can obtain information about the important parameters.

Due to skin effect we have two important regimes in metals - normal and anomalous. The linewidth in the normal conductivity regime ($l \ll \delta$), l is the mean free path of the electron, δ is the skin depth) can be roughly given by (31)

$$\begin{aligned} \Gamma_{pp} &\approx \Gamma_{pp}^{\text{rel}} + \Gamma_{pp}^{\text{ex}} \\ &= \frac{1.45 \omega \lambda}{\gamma^2 M_s} + \frac{C A^{\frac{1}{2}} \omega^{\frac{1}{2}}}{f^2} \end{aligned} \quad (8)$$

where the first term is contributed by the magnetic relaxation damping and the latter by the exchange conductivity mechanism. It is evident from (8) that the linewidth will be dominated by exchange effects at low frequencies and by magnetic relaxation damping at high frequencies³¹

Anomalous Skin Effect Region

In the low temperature region where the average mean free path l becomes larger than the skin depth (i.e., $l > \delta$), the skin effect becomes anomalous. In this regime the effectiveness of the electrons in absorbing energy from the radiation field is reduced, or stated differently the conductivity σ becomes k (wave number of the spin wave excited) dependent and decreases for increasing k .

In the extreme anomalous skin effect region ($l \gg \delta$) the conductivity becomes proportional to k . Since the value of k at resonance is large ($\approx 10^5$) this means that the effect of anomalous conductivity will be to reduce the exchange effects, as compared to the case when the conductivity is taken equal to its $k = 0$ value (normal region) Vonsovski et al⁴³ and Turov⁵⁰ find that the linewidth becomes independent of temperature in this region

Vonsovski et al⁴³ have shown that the spin wave damping can take place only when it has sufficient energy (of the order of J_{s-d}) required to flip the conduction electron spin. The energy of a typical spin wave ($k \sim \frac{1}{\xi} \approx 10^5$) is of the order of 10^{-6} eV. This is several orders of magnitude smaller than $J_{s-d} \approx 10^{-1}$ eV found by Kasuya⁴². Therefore, the spin wave damping will not be favoured for low energy spin waves due to the large exchange gap between bands of s-electrons with different spin orientations and the energy conservation. Recently⁴⁴, however, it has been pointed out that this in itself is not such a serious difficulty because in principle non-resonant absorption of energy to non-stationary states of s-spins is possible at very low magnon energies Kittel and Mitchell⁵¹ considered the effect of s-d exchange interaction on the linewidth. They assumed that the conduction electron is relaxed to the lattice independently by a rapid mechanism. Turov⁵⁰ has shown that the linewidth depends very strongly on J_{s-d} ($\propto J_{s-d}^4$). The value of J_{s-d} is not precisely

known. However, to find agreement with experiments a value of J_{s-d} of the order of 10^{-2} eV will be required. This value is not entirely unreasonable but is an order of magnitude smaller than that found from some other measurements⁴².

Secondly to fit the frequency dependence of the linewidth a frequency dependent J_{s-d} is required which is highly unlikely. According to Vonsovskii et al⁴³ and Turov⁵⁰ the linewidth should decrease (in the normal conducting regime) with decreasing temperature in direct proportion to electrical resistivity. This does not agree with the observed behaviour in any temperature range⁴⁹. Turov⁵⁰ also considered the contribution made to the resonant linewidth by the interaction of the s-spins with the magnetic field due to the spin waves and showed that these processes made a much smaller contribution to the linewidth. Recently Heinrich et al⁴⁴ has investigated the influence of s-d exchange on relaxation of magnons in metals (Ni, Fe). By phenomenologically taking into account the finite value of the s-spin lattice relaxation time they seem to be able to account for their observations on temperature dependence of the relaxation frequency λ in iron²⁴. However, the measurements of Bhagat et al^{31,52,53} do not show any such dependence and, therefore, the applicability of their theory to iron is far from being unequivocal. They, however, admit that their theory is not likely to explain the observed linewidths in nickel. Very recently Lechner et al⁵⁴ have claimed to have been able to explain the observed linewidths

in $\text{K}_2\text{CuCl}_4 \cdot 2\text{H}_2\text{O}$ using Van Vleck's moment expressions⁵⁵

Unfortunately, it is not possible to explain the linewidths in metals (specially Ni) on the basis of existing theories.

CHAPTER - II

PHENOMENOLOGICAL THEORY OF FERROMAGNETIC RESONANCE

The results of measurements on resonance frequency and linewidth in FMR can be interpreted in terms of a macroscopic dynamic equation called the phenomenological equation^{47,48} and information about important relaxation processes in a ferromagnet can be obtained from such an equation. The phenomenological theory of FMR is thus a macroscopic theory.

2.1 Effective Internal Field in a Ferromagnet

The equation of motion for the magnetization vector \vec{M} of a ferromagnet placed in a static magnetic field B in z -direction is of the form

$$\frac{d\vec{M}}{dt} = \gamma [\vec{M} \times \vec{H}_1] \quad (2.1)$$

where $\gamma = \frac{ge}{2mc}$ is the spectroscopic splitting factor and H_1 is the effective internal magnetic field inside the ferromagnet. It includes besides the external field all internal fields arising out of magnetic anisotropy energy, demagnetizing dipolar energy and magnetostriction (state of stress). Lorentz and Weiss fields are omitted as they are parallel to \vec{H} and hence their cross product with \vec{M} will be identically zero. Equation (2.1) is the fundamental equation. We will use this to obtain the expression for the resonant frequency.

2.2 General Expression for the FMR Frequency

The FMR frequency was first obtained by Kittel³ using the phenomenological theory. The effective values of the magnetic field components inside the ferromagnet, ignoring anisotropy effects, are

$$H_{x1} = B_x - N_x M_x \quad (2.2a)$$

$$H_{y1} = -N_y M_y \quad (2.2b)$$

$$H_{z1} = B - N_z M_z \quad (2.2c)$$

where N_x , N_y and N_z are called the classical demagnetizing factors. M_x , M_y and M_z are the x, y and z components of the magnetization. The radiation field B_x is along x-direction.

Since M_x and M_y are small compared with M_z as the specimen is magnetized to saturation in the z-direction, we can safely neglect products such as $M_x M_y$ in the equations (2.1) which become

$$\frac{dM_x}{dt} = \gamma [B + (N_y - N_z)M_z] M_y \quad (2.3a)$$

$$\frac{dM_y}{dt} = \gamma [M_z B_x - (N_x - N_z)M_x M_z - M_x B] \quad (2.3b)$$

$$\frac{dM_z}{dt} \approx 0. \quad (2.3c)$$

These equations (2.3) can be solved by setting $M_x = M_x^0 e^{i\omega t}$, $M_y = M_y^0 e^{i\omega t}$ and the susceptibility $\chi_x = \frac{M_x}{B_x}$ can be found (2.4) to be $\chi_x = \frac{\chi_0}{1 - (\frac{\omega}{\omega_0})^2}$ where χ_0 is the static

susceptibility and ω_0 is the ferromagnetic resonance frequency for an ellipsoidal sample given by

$$\omega_0 = \gamma \left[\left\{ B + (N_x - N_z)M \right\} \left\{ B + (N_y - N_z)M \right\} \right]^{\frac{1}{2}} \quad (2.5)$$

Here μ_z has been replaced by μ .

2.3 Quantum Mechanical Derivation of FMR Frequency

The FMR frequency can also be derived quantum mechanically^{36,55,56} considering suitable models of the ferromagnet. One such model is the Heisenberg model with magnetic dipolar interaction first considered by Van Vleck⁵⁵. This model assumes that (a) the electrons are localized on the atoms (b) only one electron per atom is present (c) the electrons are exchange coupled through an interaction of the type $-J_{ij} \vec{S}_i \cdot \vec{S}_j$, where J_{ij} is an exchange integral, S_i and S_j are the spin operators corresponding to the electrons on the atoms at the lattice sites i and j . Apart from this exchange interaction, there is also a magnetic dipole-dipole interaction. This dipolar interaction depends not only on the orientation of the spins with each other but also on their orientations with the line joining them.

The model Hamiltonian is of the form

$$H = g \mu_B B \sum_j S_j^z + \sum_{k > j} J_{kj} \vec{S}_k \cdot \vec{S}_j + \sum_{k > j} D_{kj} \left[\vec{S}_j \cdot \vec{S}_k - \frac{3}{r_{jk}^2} (\vec{S}_j \cdot \vec{r}_{jk})(\vec{S}_k \cdot \vec{r}_{jk}) \right] \quad (2.6)$$

where the terms on the right hand side of equation (2.6) denote respectively the Zeeman energy (associated with the static magnetic field in the negative z-direction), exchange energy and the magnetic dipolar energy. The coefficients D_{kj} have the value

$$D_{kj} = \frac{g^2 \mu_B^2}{r_{jk}^3}$$

if the dipole-dipole interaction is of true magnetic origin. The values of D_{kj} larger in magnitude can also occur as a result of spin-orbit coupling. Such dipolar interaction is called pseudo-dipolar interaction and is of short range character. We do not consider such pseudo-dipolar coupling here.

We rewrite the Hamiltonian (2.6) as

$$H = g \mu_B S_J^z + \sum_{k>j} A_{kj} \vec{S}_k \cdot \vec{S}_j - 3 \sum_{k>j} D_{kj} r_{jk}^{-2} (\vec{S}_k \cdot \vec{r}_{jk})(\vec{S}_j \cdot \vec{r}_{jk}) \quad (2.6)$$

where $A_{kj} = J_{kj} + D_{kj}$

The equation of motion method is now used to obtain the resonance frequency.

The equation of motion for the atomic spin S_j can be written as follows

$$\frac{dS_j}{dt} = \frac{1}{\hbar} [H, S_j] \quad (2.7)$$

where $[H, S_j]$, denotes the commutator of S_j with the

Hamiltonian (2.6) of the spin system.

Using the well known commutation relations

$$[S_J^x, S_k^y] = i \delta_{Jk} S_J^z \quad \text{etc}$$

equation (2.7) yields for the three components of the total spin S

$$\begin{aligned} \hbar \frac{dS^x}{dt} = -g\mu_B B S^y + 3 \sum_{k \neq J} D_{kJ} (\alpha_{Jk} S_J^x + \beta_{Jk} S_J^y + \gamma_{Jk} S_J^z) \times \\ (-\beta_{Jk} S_k^z + \gamma_{Jk} S_k^y) \end{aligned} \quad (2.8a)$$

$$\begin{aligned} \hbar \frac{dS^y}{dt} = g\mu_B B S^x + 3 \sum_{k \neq J} D_{kJ} (\alpha_{Jk} S_J^x + \beta_{Jk} S_J^y + \gamma_{Jk} S_J^z) \times \\ (\alpha_{Jk} S_k^z - \gamma_{Jk} S_k^x) \end{aligned} \quad (2.8b)$$

and

$$\begin{aligned} \hbar \frac{dS^z}{dt} = 3 \sum_{k \neq J} D_{kJ} (\alpha_{Jk} S_J^x + \beta_{Jk} S_J^y + \gamma_{Jk} S_J^z) \times \\ (-\alpha_{Jk} S_k^y + \beta_{Jk} S_k^x) \end{aligned} \quad (2.8c)$$

here α_{Jk} , β_{Jk} and γ_{Jk} denote the direction cosines of the vector \vec{r}_{Jk} joining the atomic spins at the lattice sites i and J . $S = \sum_J S_J$ denotes the total spin of the system.

It has been assumed here that the skin depth is large compared to the dimensions of the specimen so that the radiation field is uniform throughout the sample. The magnetic operator S^x which is responsible for the absorption can be written as $S^x = \sum_J S_J^x$.

The exchange energy commutes with any component of the total spin and that is why it does not appear in eqn (2.8).

In the above equations (2.8) one can neglect products such as $S_j^x S_k^y$ on the basis that in a ferromagnetic medium, the specimen will be practically magnetized to saturation along the field direction and the matrix elements of S^x and S^y will be smaller than those of S^z . In the products such as $S_j^x S_k^z$, we replace S_j^z by its mean value $\frac{-\hbar}{g \mu_B N}$ where N is the number of atoms in the crystal

Using these approximations and assuming that the atoms in the crystal are symmetrically situated (so that the terms such as $\sum_j \alpha_{jk} \gamma_{jk} D_{kj}$ etc vanish), we obtain

$$\begin{aligned} \hbar \frac{dS^x}{dt} &= -g \mu_B B S^y + (N_z - N_y) g \mu_B M S^y \\ \hbar \frac{dS^y}{dt} &= g \mu_B B S^x - (N_z - N_x) g \mu_B M S^x \\ \frac{dS^z}{dt} &\approx 0 \end{aligned} \quad (2.8')$$

where the shape dependent demagnetizing factors are defined as follows

$$N_x = \frac{1}{N} \sum_k \frac{1 - 3 \alpha_{jk}^2}{r_{jk}^3} + \frac{4\pi}{3}$$

and the corresponding expressions for N_y and N_z

Eqn. (2.8) can be solved by setting

$$S^x = S_o^x e^{i\omega t} \quad \text{and} \quad S^y = S_o^y e^{i\omega t},$$

to yield the Kittel resonance frequency (2.5).

It should, however, be pointed out here that the present model is particularly suited to the case of ferromagnetic insulators such as garnets, since the localized electron spin concept is applicable in their cases. This model was employed by Van Vleck⁵⁵ to explain ferromagnetic resonance absorption and linewidth*.

2.4 Effect of the Shape of the Specimen on FMR Frequency

As we know that in a ferromagnet there are strong internal fields arising out of magnetic anisotropy⁵⁷ caused by the magnetic interactions between electrons, the effective internal field H_1 will depend on the symmetry of the crystal, the shape of the sample, the magnetization and its direction and on the direction of the external static field B in relation to the crystallographic axes. From the expression (2.5) it is clear that the resonance frequency depends on the shape of the sample as the demagnetizing factors enter into its expression. We consider some special cases of eqn. (2.5).

(a) Plane For an infinite plane sample in the x-z direction

$$N_x = N_z = 0, \quad N_y = 4\pi$$

$$\text{and } \omega_0 = \gamma [B (B + 4\pi M_z)]^{\frac{1}{2}} = \gamma (B B_1)^{\frac{1}{2}}$$

where B_1 denotes the magnetic induction. We, therefore, notice that ω_0 is equal to the Larmor frequency for a fictitious field $(B B_1)^{\frac{1}{2}}$.

(b) Sphere. For a spherical sample all the demagnetizing factors will be equal, i.e.,

*The linewidth is defined as the width of the resonance curve at the half power absorption.

$$N_x = N_y = N_z = 4\pi/3$$

and $\omega_0 = \gamma B$

i.e., for a spherical sample ω_0 is equal to the Larmor frequency.

2.5 Magnetic Anisotropy and its Influence on FMR Frequency

Ferromagnetic crystals have "easy" and "hard" directions of magnetization, i.e., it is easy to magnetize a crystal in a direction (called easy direction) than in the other (called hard direction). The excess amount of energy needed to magnetize a crystal in a "hard" direction than in an "easy" direction is called the magnetic anisotropy energy⁵⁷.

In a crystal there is an intense, inhomogeneous and asymmetric electric field (called the crystal field) produced by the constituent ions⁵⁸. As a result of the crystal field and the spin-orbit coupling, the electron orbits are distorted along particular directions and the magnetic fields associated with these orbits have directional properties and the spin-magnetic moment orients itself along the resultant of the fields produced by the orbits and thus anisotropy originates. The spin-orbit coupling is a consequence of the partial quenching of the orbital angular momentum by the crystal field. What happens in an asymmetric electric field as exists in a crystal is that the orbital angular momentum components are no longer constant (in a symmetric field one component usually taken as L_z and the square of the total orbital angular

momentum L^2 are constants of the motion) In a crystal L_z will no longer be a constant of the motion, although to a good approximation L^2 may be a constant of the motion When L_z averages to zero, the orbital angular momentum is said to be completely quenched However, the quenching is usually not complete^{57,58} and, therefore, the spin interacts with the orbital motion which in turn interacts with the crystal lattice by means of the electrostatic field. Thus the spin (or the magnetization) "sees" the crystal lattice and its geometry This is why the anisotropy depends on the crystal symmetry.

Owing to the spin-orbit coupling, the spin vectors responsible for ferromagnetism feel slightly anisotropic electrostatic forces which connect the orbital angular momenta of different atoms and whose bonding effect is called orbital valence⁵⁹ Consequently, anisotropic dipole-dipole and anisotropic quadrupolar couplings (usually called pseudo-dipolar and pseudo-quadrupolar interactions respectively) are produced. These interactions are used to explain magnetic anisotropy in ferromagnets. Van Vleck was the first to introduce pseudo-dipolar interaction to explain anisotropy of single cubic metal crystals. As will be mentioned later, Van Vleck also considered a microscopic model of anisotropy with a quadrupolar coupling between the spins to explain resonance linewidth and frequency shift.

For cubic crystals anisotropy energy can be written in the form

$$E_{an} = K_1(\alpha^2 \beta^2 + \beta^2 \gamma^2 + \gamma^2 \alpha^2) + K_2 \alpha^2 \beta^2 \gamma^2 \quad (2.9)$$

where K_1 and K_2 are the first and second order anisotropy constants respectively and α , β and γ denote the direction cosines of the magnetization referred to the crystal axes. The constants K_1 and K_2 can be determined from experiment for iron at room temperature

$$K_1 = 4.2 \times 10^5 \text{ erg/cm}^3, \quad K_2 = 1.5 \times 10^5 \text{ erg/cm}^3 \quad (2.10)$$

For crystals with a single preferred axis, such as cobalt, the anisotropy energy may be of the form

$$E_{an} = K_1 \sin^2 \phi + K_2 \sin^4 \phi$$

where ϕ is the angle between the magnetization and the easy axis, higher order terms are usually neglected. For Cobalt at room temperature

$$K_1 = 4.1 \times 10^6 \text{ erg/cm}^3, \quad K_2 = 1.0 \times 10^6 \text{ erg/cm}^3 \quad (2.11)$$

The anisotropy constants depend strongly on temperature

The anisotropy energy influences FMR frequency through its dependence on the angles formed by the static field with the crystal axes. At a fixed radiation field frequency the value of the constant magnetic field at which the resonance occurs, will be lowest near the "easy" direction and highest near the "hard" direction. This angular dependence has been confirmed by the experiments of Kip and Arnold⁶⁰.

Kittel³ considered the effect of the macroscopic anisotropy energy of the form (2.9) and found that its effect can be included by the proper alteration of the demagnetizing factors used in (2.5). Van Vleck⁵⁵, however, considered the microscopic model of anisotropy with a quadrupolar coupling between the spins of the type

$$\sum_{\substack{k \\ k \neq j}} K_{jk} r_{jk}^{-4} (\vec{S}_j \cdot \vec{r}_{jk})^2 (\vec{S}_k \cdot \vec{r}_{jk})^2 \quad (2.12)$$

It should be noticed that for identical experimental situations both Kittel and Van Vleck get almost identical results.

2.6 Effect of Domain Structure

In deriving (2.5) it has been assumed that the static magnetic field is large enough to break down the domain structure and the specimen can be regarded as a single domain magnetized to saturation so that the anisotropy effects might be neglected. In the case of weaker magnetizing fields there will be a multidomain structure in the specimen as a consequence of which additional peaks will appear in the absorption spectrum.

Nagamiya⁶¹ who calculated the effect of domain structure on FMR in single crystals with tetragonal symmetry has explained the additional peaks observed experimentally by Bickford⁶².

2.7 Phenomenological Approach to Damping

The divergence of dynamic susceptibility χ_x to ω at resonance ($\omega = \omega_0$) means only that attenuation (damping) processes have not been taken into consideration. It has been

pointed out in the preceding chapter that a part of the width of the ferromagnetic resonance line arises out of the interaction of the uniform mode with other modes such as phonons, magnons and conduction electrons of the system. The effect of these interactions can be taken care of formally in the phenomenological theory by adding to the right hand side of the eqn (2.1) the relaxation term which describes damping

The first phenomenological equation was suggested by Landau-Lifshitz⁴⁷ in 1935 and has the following form

$$\frac{d\vec{M}}{dt} = -\gamma'(\vec{M} \times \vec{B}) - \frac{\lambda}{M} \vec{M} \times (\vec{r} \times \vec{B}) \quad (2.13)$$

where λ is the relaxation (damping) parameter

A slightly different way of writing this equation was suggested by Gilbert⁴⁸ whose equation reads

$$\frac{d\vec{M}}{dt} = -\gamma_G (\vec{M} \times \vec{B}) + \lambda_G (\vec{M} \times \frac{d\vec{M}}{dt}) \quad (2.14)$$

The basis of writing it in this form for the relaxation term in (2.13) and (2.14) is that it represents the torque which characterizes the damping of the magnetization vector \vec{M} to B along a spiral path.

Another phenomenological equation was introduced by Bloch to describe nuclear magnetic resonance and was employed by Bloembergen to explain ferromagnetic relaxation. The Bloch-Bloembergen⁶³ phenomenological equations read

$$\begin{aligned} \frac{dM_{x,y}}{dt} &= -\gamma'(\vec{r} \times \vec{B})_{x,y} - \frac{M_{x,y}}{T_2} \\ \frac{dM_z}{dt} &= -\gamma'(\vec{M} \times \vec{B})_z - \frac{M_z - M_s}{T_1} \end{aligned} \quad (2.15)$$

where τ_1 and τ_2 are relaxation parameters M_s is the saturation magnetization. This form of the relaxation term characterizes the exponential decay of the transverse components of magnetization to B with a characteristic time constant τ_2 .

For weak radiation fields when we put $M_z = M_s$, the Landau-Lifshitz-Gilbert and Bloch-Bloembergen equations are equivalent. The shape of the absorption line in all these cases is Lorentzian.

Yager et al.⁶⁴ studied the ferromagnetic resonance in single crystals (spherical samples) of nickel ferrite ($\text{NiO} \cdot \text{Fe}_2\text{O}_3$) at room temperature at 24 KMc/sec. The value of the g -factor obtained by them was 2.19 and the first order magnetic anisotropy constant K_1 was found to be $-6.27 \cdot 10^4$ erg/cm³ as compared to $-6.2 \cdot 10^4 \pm 10\%$ observed from hysteresis loop measurements by Galt et al.⁶⁵. They employed Landau-Lifshitz damping and were able to fit the resonance curve quite satisfactorily.

A similar FMR study on small spherical samples of nickel ferrite was made by Healy⁶⁶ over a wide temperature range between -195°C to 588°C . The g -factor and first and second order anisotropy constants were obtained from the study of the variation of the linewidth with temperature. The g -value was found to be essentially constant over the entire temperature range. The anisotropy constants K_1 and K_2 showed different sign behaviour over the temperature range. Healy was, however, able to fit his resonance curves by either type of damping - Landau-Lifshitz or Bloch type. His results showed that the

linewidth decreases with the increase in temperature which suggests that the spin-lattice and dipolar interactions do not contribute materially to the linewidth. The measurements on spin-lattice relaxation times made by Damon⁶⁷ have indicated that the spin-lattice interaction contributes little to the linewidth. The conclusion drawn by Healy that the spin-lattice relaxation does not contribute to the line broadening is not totally correct as the relaxation time occurs in the phenomenological equation as a gross effect and cannot be separated in a simple manner.

Young et al⁶⁸ who investigated FMR in nickel and supermalloy in two different conditions of magnetization, namely, (1) the applied magnetic field is parallel to the surface of the sample and (2) it is perpendicular to the surface, found that the shape of the curve was correctly predicted only in the case of parallel magnetization and in the case of perpendicular magnetization there were marked discrepancies (in the low field region) such as the appearance of a second minimum in the frequency shift curve. This they tried to explain on the basis that in the perpendicular case they were in the region of unsaturated magnetization and the theory is consequently unable to explain the phenomenon. They found that though both the Bloch and Landau-Lifshitz damping terms lead to Lorentz shape of the curve, the former was more satisfactory than the latter.

According to the usual theory of FMR, the ratio of absorptions in the two cases of parallel and perpendicular

magnetizations is independent of the numerical value of the damping constant but its dependence on the parameter $\frac{2\pi\gamma M_s}{\omega_0}$ is different in the two cases. Their results⁶⁸ differed from the predicted ratio in the most favourable case (parallel magnetization case) by a factor of about 1.5, and they were led to interpret that neither type of damping term was appropriate.

Ament and Rado⁶⁹ gave an electromagnetic theory of "spin wave resonance" experiment and satisfactorily explained the exchange shift as well as the shape of the absorption line. They used Landau-Lifshitz type of damping in their calculation.

Rado and Weertman⁷⁰, who studied spin wave resonance in single and polycrystals of nickel iron alloy having extremely low magnetic anisotropy at 3 and 4 KMc/sec, showed that the resonance curves could be interpreted on the basis of the theory of Ament and Rado⁶⁹ by using an effective exchange stiffness constant A of $(3.3 \pm 0.5) \times 10^{-6}$ erg/cm and a spectroscopic g factor of 2.06 ± 0.01 . They found, however, that the agreement (qualitative) could be destroyed if a Landau-Lifshitz or Bloch damping term were included.

Experimental data taken at liquid nitrogen temperature showed an increase in the resonance linewidth and a decrease in the resonance field. A part of the discrepancy was attributed to the anomalous effects caused by the electronic mean free path. More recently Hirst and Irwin⁷¹ have shown that the discrepancies encountered by Rado and Weertman⁷⁰ in trying to fit their nitrogen temperature data cannot be attributed to the

non-local conductivity (anomalous skin depth effect).

The destroying of the agreement by using either type of damping (Landau-Lifshitz or Bloch) is not very surprising. Rado and Weertman⁷⁰ studied the effect of exchange on the linewidth, and their experimental conditions were such that the exchange term was predominant and the relaxation term was made negligibly small by suitably choosing a specimen of zero anisotropy, of large conductivity and by working at low field. Therefore, addition of any damping term will spoil the agreement with the experiment, as the relaxation processes were not operating.

The ferromagnetic resonance linewidth in a metal³¹ is believed to be due to (1) magnetic relaxation damping whose microscopic origin is not exactly known. It is described by including Landau-Lifshitz or Bloch-Bloembergen type of damping terms in the equation of motion for magnetization and (2) exchange conductivity broadening which arises due to the fact that the exchange stiffness opposes the tendency of the dynamic magnetization to vary in the skin depth. It depends upon the exchange stiffness constant, conductivity and magnetic anisotropy that might be present. It has already been pointed out in the preceding chapter that the linewidth due to exchange conductivity broadening varies as $\omega^{\frac{1}{2}}$ (where ω is the frequency) and that due to magnetic relaxation damping as ω . The measurements on FMR linewidths in nickel and iron have been made by many workers³³⁻³⁵. These authors have used

Landau-Lifshitz type of damping. Their results are not all in agreement with each other. Salamon³⁴ found that the value of the g-factor in the case of nickel suddenly increases as he goes from below T_c to above T_c (T_c being the Curie temperature), while Bhagat et al³⁵ observed that the increase in the g-factor was rather gradual. The reasons for this discrepancy are not known. It is possible, however, that the spin-orbit coupling effects might be very important in the case of nickel. The band structure effects of the d-electrons and the influence of the conduction electrons on them should also be taken into consideration. Very recently Kambersky⁷² has studied FMR in iron whiskers and has reported linewidths at the highest frequency (73GHz). The linewidths found by Bhagat et al^{52,53} at the corresponding temperatures and frequencies (upto 35 GHz) are larger than those obtained by Kambersky.

2.8 Discussion

On the basis of the interpretation of the results of measurements on FMR using phenomenological theory, one can say that the theory explains the shape of the resonance line and also the anisotropy satisfactorily. The linewidth and its temperature dependence is not, however, explained by the theory adequately. The reason is probably that as absorption is a gross effect and the relaxation terms used also represent an overall effect due to all the damping mechanisms present in the system, and so the relaxation parameter can be chosen to

fit the resonance curve nicely, but the details of the line cannot be reproduced by such phenomenological parameter whose microscopic origin is not exactly understood

We, therefore, conclude by saying that the phenomenological theory is not adequate for a quantitative description of FMR linewidths.

CHAPTER - III

GREEN FUNCTION THEORY OF FMR AND MAGNON-PHONON COUPLING3.1 Green Function Formalism

Solid state theory problems are inherently many-body problems. The usual quantum mechanical perturbation theory methods are inappropriate to treat these many-body problems. However, in recent years, there has been a considerable interest in trying to treat these problems by the methods of quantum field theory. One of these methods is the method of double time temperature dependent Green functions, which has been applied very extensively and successfully to a wide range of problems of solid state physics such as ferromagnetism, superconductivity and lattice dynamics, etc. The method is non-perturbative. The poles of the Fourier transform of the Green function yield information regarding the elementary excitations of the solid and their decays. Most of the quantities of immediate physical interest can be obtained in terms of these Green functions.

We can define different kinds of Green functions. The double time causal Greenfunction defined in terms of the average value of the T-product of operators, or the retarded and advanced Green functions. Throughout the present work, we shall use retarded Green functions. There exist several reviews⁷³ and texts⁷⁴ on Green functions. We would closely follow an excellent review by Zubarev⁷³. The retarded Green

function is defined as follows

$$G_{AB}(t, t') = -i\theta(t-t') \langle [A(t), B(t')] \rangle = \langle\langle A(t), B(t') \rangle\rangle \quad (3.1)$$

where A and B are Heisenberg operators

$$A, B = e^{iHt} A, B e^{-iHt}$$

H is the system Hamiltonian

$$\begin{aligned} \theta(t) &= 1 && \text{for } t > 0 \\ &= 0 && \text{otherwise} \end{aligned}$$

(a system of units in which $\hbar = 1$ is used)

The bracket $[,]$ denotes the commutator and $\langle - \rangle$ denotes the grand ensemble average, namely, for any operator O_p

$$O_p = T_r (e^{-\beta H} O_p) / T_r e^{-\beta H}$$

Here T_r denotes the trace of the quantity that follows

$\beta = (k_B T)^{-1}$, k_B being the Boltzman constant and T is the temperature of the system. The operators $A(t)$ and $B(t')$ satisfy the equation of motion

$$\frac{1dA}{dt} = [A, H] \quad (3.2)$$

Differentiating the Green function (3.1) with respect to time we obtain

$$\frac{1dG(t-t')}{dt} = \delta(t-t') \langle [A(t), B(t')] \rangle + \langle\langle [A(t), H], B(t') \rangle\rangle \quad (3.3)$$

The Green function $\langle\langle [A(t), H], B(t') \rangle\rangle$ in eqn. (3.3) is

general involves Green functions of higher order than the original Green function $\llbracket A(t), B(t') \rrbracket$, except, of course, for the cases of non-interacting systems where exact solution can be obtained. One is, therefore, forced to linearize the eqn (3.3) by suitable decoupling approximations

The time correlation functions which are of physical interest are defined as

$$F_{BA}(t, t') = \langle B(t') A(t) \rangle, \quad F_{AB}(t, t') = \langle A(t) B(t') \rangle \quad (3.4)$$

In the statistical equilibrium, these functions F_{BA} and F_{AB} depend, as do the Green functions, only on the difference $t-t'$. Equal time correlation functions are called auto-correlation functions and they give the average values of the products of operators. One can express the correlation functions in terms of the spectral density function $J(\omega)$ as

$$F_{BA}(t-t') = \int_{-\infty}^{\infty} d\omega J(\omega) e^{-i\omega(t-t')} \quad (3.5a)$$

$$F_{AB}(t-t') = \int_{-\infty}^{\infty} d\omega J(\omega) e^{\beta\omega} e^{-i\omega(t-t')} \quad (3.5b)$$

Since $G(t-t')$ is the difference of F_{AB} and F_{BA} , it follows that

$$G(\omega + i\varepsilon) - G(\omega - i\varepsilon) = -i(e^{\beta\omega} - 1) J(\omega) \quad (3.6)$$

$$\varepsilon \rightarrow 0.$$

Here $G(\omega)$ is the Fourier transform of $G(t-t')$ defined as follows

$$G(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\omega(t-t')} G(t-t') d(t-t') \quad (3.7)$$

The expression (3.6) relating the Green-function and the spectral density function is important. Once the retarded Green function is known, the spectral density function can be obtained and then one can get various correlation functions at all times and at all temperatures.

3.2 Linear Response Theory

Kubo⁷⁵ has given an elegant formulation of the problem of the response of a quantum mechanical system to an external perturbation. The theory assumes that the response of the system to the external influence is linear. The higher or the nonlinear terms are neglected in the above analysis, but they are important in the theory of phenomena which arise due to nonlinearity.

Let us consider a system with time-independent Hamiltonian H_0 . A perturbation H_{int}^t which depends on time explicitly has been switched on at $t = -\infty$, when the system was in thermal equilibrium. Now the system is described by the Hamiltonian

$$H = H_0 + H_{int}^t \quad (3.8)$$

The ensemble average of any operator A is given by

$$\langle A \rangle = \text{Tr} [\rho(t)A] \quad (3.9)$$

where $\rho(t)$ is the density matrix representing the ensemble and satisfies the following equation

$$i \frac{d\rho(t)}{dt} = [H, \rho(t)] = [H_0 + H_{int}^t, \rho(t)] \quad (3.10)$$

Let $\rho(t) = \rho_0 + \Delta\rho(t)$ such that

$$\Delta\rho(t) = 0 \quad \text{for } t = -\infty \quad \text{and} \quad \rho(-\infty) = \rho_0 = \frac{e^{-\beta H}}{\text{Tr } e^{-\beta H}} \quad (3.11)$$

Restricting to the linear term in the external perturbation and making use of the initial conditions (3.11), the change in the density matrix can be obtained as

$$\Delta\rho(t) = -i \int_{-\infty}^t e^{iH(t-t')} [H_{int}^t, \rho] e^{-iH(t-t')} dt' \quad (3.12)$$

Now the average value of the operator A is given by

$$\begin{aligned} A &= \text{Tr} \left[\left\{ \rho_0 + \Delta\rho(t) \right\} A \right] \\ &= A_0 + i \int_{-\infty}^{\infty} \ll A(t), H_{int}^{t'} \gg dt' \end{aligned} \quad (3.13)$$

Considering $H_{int}^{t'} = F(t') O$, where $F(t')$ is the force coupled to the operator part O of the dynamical system. Then

$$H_{int}^{t'} = F(t') e^{iH_0 t'} O e^{-iH_0 t'} = F(t') O(t') \quad (3.14)$$

Then the effect $\Delta A = A - A_0$ will be given by

$$\Delta A = \int_{-\infty}^{\infty} G(t-t') F(t') dt' \quad (3.15)$$

Thus the retarded Green function is essentially a response function.

3.3 Dynamic Magnetic Susceptibility

In studying ferromagnetic resonance one has to find the changes in the average magnetization components $\Delta m_1(t)$ ($1 = x, y$) caused by the radiation field $B_1(t)$, as they yield us dynamical magnetic susceptibility

$$\Delta m(t) = \chi B(t) \quad (3.16)$$

which determines the transfer of energy from the radio frequency field to the spin system. Remembering that

$$\Delta m_1 = \mu_B \Delta S_f^1(t) \quad (3.17)$$

here \vec{f} refers to the lattice site. One can see that the problem reduces to evaluating the average changes in the spin operators $\Delta S_f^1(t)$, which is independent of \vec{f} because of translational invariance.

The Hamiltonian of a ferromagnetic system in the presence of a radiation field can be written in the form

$$H = H_0 + H_{int}^t \quad (3.18)$$

Here H_{int}^t describes the interaction of the spin system and the radiation field, and H_0 includes the interaction with external static magnetic field and all other interactions such as exchange interactions and dipolar interactions, etc.

Assuming that the constant magnetic field is directed along the z-axis and the oscillating field lies in the x-y plane and is uniform throughout the sample, we can write for H_{int}^t

$$H_{\text{int}}^t = -\mu_B \sum_{J, \vec{f}} B_J(t) S_{\vec{f}}^J(t), \quad (J = x, y) \quad (3.19)$$

It is obvious that in the case of a plane polarized radio frequency field and in the linear approximation, we can write for χ the following expression

$$\chi_{1j} = -2\mu_B^2 \sum_{\vec{f}} \ll S_{\vec{g}}^1, S_{\vec{f}}^j \gg_{\omega} \quad (3.20)$$

Thus to evaluate the dynamical magnetic susceptibility we must find the Fourier transform of the Green function $\ll S_{\vec{g}}^1, S_{\vec{f}}^j \gg$.

3.4 Hamiltonian of the System An Application of Green Function Method

Consider a ferromagnet in a constant magnetic field B in z -direction. The Hamiltonian⁵⁶ can be written in the form

$$H = H_z + H_{\text{ex}} + H_d \quad (3.21)$$

$$\text{where } H_z = -\mu_B B \sum_{\vec{f}} S_{\vec{f}}^z \quad (3.21a)$$

$$H_{\text{ex}} = -\frac{1}{2} \sum_{\vec{f}_1 \neq \vec{f}_2} J(\vec{f}_1 - \vec{f}_2) \vec{S}_{\vec{f}_1} \cdot \vec{S}_{\vec{f}_2} \quad (3.21b)$$

$$\text{and } H_d = \frac{1}{2N} \sum_1 N_1 M_1^2, \quad (1 = x, y, z) \quad (3.21c)$$

Here all the symbols have their usual meaning $J(\vec{f}_1 - \vec{f}_2)$ is the exchange integral which is positive, M_1 denotes the 1^{th} component of the magnetization and N_1 is the demagnetization factor.

The three terms in the Hamiltonian (3.21) respectively denote Zeeman energy, Heisenberg exchange energy and classical demagnetizing energy. The demagnetizing energy (3.21c) arises due to the finiteness of the sample dimensions and, therefore,

takes into account the demagnetization effects of the surface of the finite sample.

The demagnetizing energy (3.21c) can be written in the quantum-mechanical form using the relation

$$M_1 = \mu_B \sum_{\vec{f}} S_{\vec{f}}^1 \quad (3.22)$$

It will be convenient to change over from the spin operators to Pauli operators

$$S_{\vec{f}}^x = C_{\vec{f}} + C_{\vec{f}}^+, \quad S_{\vec{f}}^y = i(C_{\vec{f}}^+ - C_{\vec{f}}), \quad S_{\vec{f}}^z = 1 - 2n_{\vec{f}}, \quad n_{\vec{f}} = C_{\vec{f}}^+ C_{\vec{f}} \quad (3.23)$$

where $C_{\vec{f}}$ and $C_{\vec{f}}^+$ are annihilation and creation operators for spin deviation at the \vec{f}^{th} lattice site. These operators obey following commutation relations

$$C_{\vec{f}} C_{\vec{f}}^+ - C_{\vec{f}}^+ C_{\vec{f}} = 0, \quad C_{\vec{f}} C_{\vec{f}'} - C_{\vec{f}'} C_{\vec{f}} = 0, \quad \vec{f} \neq \vec{f}' \quad (3.24)$$

$$C_{\vec{f}} C_{\vec{f}}^+ + C_{\vec{f}}^+ C_{\vec{f}} = 1, \quad C_{\vec{f}} C_{\vec{f}'} = C_{\vec{f}}^+ C_{\vec{f}'}^+ = 0$$

Using (3.22) and (3.23) the Hamiltonian (3.21) can be written in the following form

$$\begin{aligned} H = & -\frac{1}{2} N \tilde{J}(0) - N \mu_B B + \left[2 \mu_B B + 2 \tilde{J}(0) \right] \sum_{\vec{f}} n_{\vec{f}} \\ & - 2 \sum_{\vec{f}_1 \vec{f}_2} J(\vec{f}_1 - \vec{f}_2) C_{\vec{f}_1}^+ C_{\vec{f}_2} - 2 \sum_{\vec{f}_1 \vec{f}_2} J(\vec{f}_1 - \vec{f}_2) n_{\vec{f}_1} n_{\vec{f}_2} \\ & + \frac{1}{2} N N_z \mu_B^2 - 2 \mu_B^2 N_z \sum_{\vec{f}} n_{\vec{f}} + \frac{2 \mu_B^2 N_z}{N} \sum_{\vec{f}_1 \vec{f}_2} n_{\vec{f}_1} n_{\vec{f}_2} \\ & + \frac{\mu_B^2}{2N} (N_x - N_y) \sum_{\vec{f}_1 \vec{f}_2} \left[C_{\vec{f}_1} C_{\vec{f}_2} + C_{\vec{f}_1}^+ C_{\vec{f}_2}^+ \right] \\ & + \frac{\mu_B^2}{N} (N_x + N_y) \sum_{\vec{f}_1 \vec{f}_2} C_{\vec{f}_1}^+ C_{\vec{f}_2} \end{aligned} \quad (3.25)$$

where $\tilde{J}(k) = \sum_{\vec{f}} J(\vec{f}) e^{i\vec{k} \cdot \vec{f}}$, $J(0) = 0$
 is the Fourier transform of $J(\vec{f})$

From equations (3.20) and (3.23) we obtain

$$\chi_{xx} = \mu_B^2 \sum_{\vec{f}} \left[\langle\langle c_{\vec{g}}, c_{\vec{f}}^+ \rangle\rangle + \langle\langle c_{\vec{g}}^+, c_{\vec{f}} \rangle\rangle + \langle\langle c_{\vec{g}}^+, c_{\vec{f}}^+ \rangle\rangle + \langle\langle c_{\vec{g}}, c_{\vec{f}} \rangle\rangle \right] \quad (3.26)$$

To determine χ_{xx} , we should evaluate the Green functions appearing in (3.26). Using eqn (3.1), we define the Green function $G_{\vec{g}\vec{f}}^+$ through

$$G_{\vec{g}\vec{f}}^+ = \langle\langle c_{\vec{g}}, c_{\vec{f}}^+ \rangle\rangle \quad (3.27)$$

and write down its equation of motion

$$\begin{aligned} i \frac{dG_{\vec{g}\vec{f}}^+}{dt} = & \sigma \delta_{\vec{g}\vec{f}} \delta(t-t') + \left[2\mu_B B - 2\mu_B^2 N_z + 2\tilde{J}(0) \right] G_{\vec{g}\vec{f}}^+ \\ & - \sum_{\vec{f}} \left[2J(\vec{g}-\vec{p}) - \frac{\mu_B^2}{N} (N_x + N_y) \right] G_{\vec{p}\vec{f}}^+ + 2 \sum_{\vec{p}} \left[2J(\vec{g}-\vec{p}) \right. \\ & - \frac{\mu_B^2}{N} (N_x + N_y) \left. \right] \langle\langle n_{\vec{g}} c_{\vec{p}}, c_{\vec{f}}^+ \rangle\rangle - 2 \sum_{\vec{p}} \left[2J(\vec{g}-\vec{p}) \right. \\ & - \frac{2\mu_B^2}{N} N_z \left. \right] \langle\langle n_{\vec{p}} c_{\vec{g}}, c_{\vec{f}}^+ \rangle\rangle \\ & + \frac{\mu_B^2}{N} (N_x - N_y) \sum_{\vec{p}} \langle\langle (1-2n_{\vec{g}}) c_{\vec{p}}^+, c_{\vec{f}}^+ \rangle\rangle \end{aligned} \quad (3.28)$$

where $\sigma = 1 - 2 \langle n_{\vec{g}} \rangle$ is the relative magnetization

This equation shows that higher order Green functions have made their appearance as usual. To write down these higher order Green functions in terms of the original ones, we

use the following decoupling procedure⁷³

$$\begin{aligned}\langle\langle n_{\vec{g}} C_{\vec{p}}, C_{\vec{f}}^+ \rangle\rangle &\approx \langle n_{\vec{g}} \rangle \langle\langle C_{\vec{p}}, C_{\vec{f}}^+ \rangle\rangle = \bar{n} G_{\vec{p}\vec{f}} \\ \langle\langle n_{\vec{g}} C_{\vec{p}}^+, C_{\vec{f}}^+ \rangle\rangle &\approx \langle n_{\vec{g}} \rangle \langle\langle C_{\vec{p}}^+, C_{\vec{f}}^+ \rangle\rangle = \bar{n} F_{\vec{p}\vec{f}}\end{aligned}\quad (3.29)$$

where $F_{\vec{p}\vec{f}} = \langle\langle C_{\vec{p}}^+, C_{\vec{f}}^+ \rangle\rangle$

With the above decoupling approximations (3.29) the equation of motion of the Green function $G_{\vec{g}\vec{f}}$ reduces to

$$\begin{aligned}i \frac{dG_{\vec{g}\vec{f}}}{dt} &= \tau \delta_{\vec{g}\vec{f}} \delta(t-t') + \frac{\sigma \mu_B^2}{N} (N_x - N_y) \sum_{\vec{p}} F_{\vec{p}\vec{f}} \\ &+ \left[2\mu_B B + \tau \right] 2\tilde{J}(0) - 2\mu_B^2 N_z \left\{ \right\} G_{\vec{g}\vec{f}} \\ &- \sum_{\vec{p}} \tau \left[2J(\vec{g} - \vec{p}) - \frac{\mu_B^2}{N} (N_x + N_y) \right] G_{\vec{p}\vec{f}}\end{aligned}\quad (3.30)$$

In the same approximation the equation of motion for $F_{\vec{g}\vec{f}}$ can be written in the form

$$\begin{aligned}i \frac{dF_{\vec{g}\vec{f}}}{dt} &= - \frac{\sigma \mu_B^2}{N} (N_x - N_y) \sum_{\vec{p}} G_{\vec{p}\vec{f}} - \left[2\mu_B B + \tau \right] 2\tilde{J}(0) \\ &- 2\mu_B^2 N_z \left\{ \right\} F_{\vec{g}\vec{f}} + \sum_{\vec{p}} \tau \left[2J(\vec{g} - \vec{p}) - \frac{\mu_B^2}{N} (N_x + N_y) \right] F_{\vec{p}\vec{f}}\end{aligned}\quad (3.31)$$

Introducing the following

$$G_{\vec{g}\vec{f}}(t-t') = \frac{1}{N} \sum_{\vec{k}} e^{i(\vec{g}-\vec{f}) \cdot \vec{k}} \int_{-\infty}^{\infty} G_{\vec{k}}(\omega) e^{-i\omega(t-t')} d\omega \quad (3.32)$$

and

$$F_{\vec{g}\vec{f}}(t-t') = \frac{1}{N} \sum_{\vec{k}} e^{i(\vec{g}-\vec{f}) \cdot \vec{k}} \int_{-\infty}^{\infty} F_{\vec{k}}(\omega) e^{-i\omega(t-t')} d\omega$$

Using eqns (3.32), (3.30) and (3.31) we obtain the following set of equations for the Fourier transforms of G and F functions

$$(\omega - \epsilon_{\vec{k}}) G_{\vec{k}}(\omega) - \delta_{\vec{k},0} \sigma \mu_B^2 (N_x - N_y) F_{\vec{k}}(\omega) = \frac{\sigma}{2\pi} \quad (3.33)$$

$$\delta_{\vec{k},0} \sigma \mu_B^2 (N_x - N_y) G_{\vec{k}}(\omega) + (\omega + \epsilon_{\vec{k}}) F_{\vec{k}}(\omega) = 0$$

$$\begin{aligned} \text{where } \epsilon_{\vec{k}} &= 2\mu_B (B - \sigma \mu_B N_z) + \sigma (2\tilde{J}(0) - 2\tilde{J}(\vec{k})) \\ &+ \sigma \mu_B^2 (N_x + N_y) \delta_{\vec{k},0} \end{aligned} \quad (3.34)$$

We shall solve eqn (3.33) separately for the case $\vec{k} \neq 0$ and the case $\vec{k} = 0$.

Case 1 $\vec{k} \neq 0$

If $\vec{k} \neq 0$ we get from eqn (3.33)

$$(\omega - \epsilon_{\vec{k}}) G_{\vec{k}}(\omega) = \frac{\sigma}{2\pi} \quad \text{i.e., } G_{\vec{k}}(\omega) = \frac{\sigma}{2\pi(\omega - \epsilon_{\vec{k}})}, F_{\vec{k}}(\omega) = 0 \quad (3.35)$$

and

$$\epsilon_{\vec{k}} = 2\mu_B (B - \sigma \mu_B N_z) + 2\sigma \{ \tilde{J}(0) - \tilde{J}(\vec{k}) \}$$

Here $\epsilon_{\vec{k}}$ is the energy of an elementary excitation in a ferromagnetic: "spin wave energy". Because of the factor σ , the elementary excitation energy $\epsilon_{\vec{k}}$ depends on the temperature.

We can use $G_{\vec{k}}(\omega)$ to evaluate σ . We know that

$$\bar{n} = \langle n_{\vec{g}} \rangle = \langle C_{\vec{g}}^+ C_{\vec{g}} \rangle$$

Now using the spectral density relation

$$\bar{n} = \frac{1}{N} \sum_{\vec{k}} \int_{-\infty}^{\infty} \frac{1}{e^{\beta \omega} - 1} \left[G_{\vec{k}}(\omega + i\varepsilon) - G_{\vec{k}}(\omega - i\varepsilon) \right] d\omega \quad (3.36)$$

and eqns (3.35) and (3.36) we obtain

$$\bar{n} = \frac{1}{N} \sum_{\vec{k}} \frac{\sigma}{e^{\beta \varepsilon_{\vec{k}}} - 1}$$

From this we got

$$\frac{1}{\sigma} = \frac{1}{N} \sum_{\vec{k}} \coth \left(\frac{\beta \varepsilon_{\vec{k}}}{2} \right) \quad (3.37)$$

The quantity $\varepsilon_{\vec{k}}$, as pointed out earlier, determines the elementary excitation spectrum in the system and cannot be negative (if we assume that $\varepsilon_{\vec{k}}$ can take on negative values, we are led to a contradiction: $n < 0$). The solution (3.35) is thus possible only if $\varepsilon_{\vec{k}} \geq 0$ for all \vec{k} . As the second term in $\varepsilon_{\vec{k}}$ is non-negative, we can get the solution considered here only in sufficiently high fields

$$B - \sigma \mu_B N_z \geq 0 \quad (3.38)$$

which means that the external static magnetic field must "overcome" the demagnetizing influence of the boundaries, i.e., the ferromagnet should behave as a single domain.

Case 2

$$\vec{k} = 0$$

For $\vec{k} = 0$, we get from eqn (3.33)

$$\begin{aligned} (\omega - \varepsilon_0) G_0(\omega) - \sigma \mu_B^2 (N_x - N_y) F_0(\omega) &= \frac{\sigma}{2\pi} \\ \sigma \mu_B^2 (N_x - N_y) G_0(\omega) + (\omega + \varepsilon_0) F_0(\omega) &= 0 \end{aligned} \quad (3.39)$$

$$\text{where } \mathcal{E}_0 = 2\mu_B B + \sigma \mu_B^2 (N_x + N_y - 2N_z) \quad (3.40)$$

From the above equations, we get

$$\begin{aligned} G_0(\omega) &= \frac{\sigma}{2\pi} \frac{\omega + \mathcal{E}_0}{\omega^2 - \omega_0^2} \\ F_0(\omega) &= -\frac{\sigma}{2\pi} \frac{\mu_B^2 (N_x - N_y)}{\omega^2 - \omega_0^2} \end{aligned} \quad (3.41)$$

$$\text{where } \omega_0 = \sqrt{A_1 A_2} = 2\mu_B \left[\left\{ B + \sigma \mu_B (N_x - N_y) \right\} \left\{ B + \sigma \mu_B (N_y - N_z) \right\} \right]^{\frac{1}{2}}$$

This is called the resonant frequency for the uniform precession mode and was first obtained by Tyablikov⁵⁶.

Using the relation

$$\ll A, B \gg_{\omega}^* = - \ll A^+, B^+ \gg_{\omega} \quad (3.42)$$

and eqns (3.26) and (3.41) we can write down the following expressions for the real and imaginary components of the dynamical magnetic susceptibility χ_{xx} .

$$\chi'_{xx} = \chi_0 (A_2/A_1)^{\frac{1}{2}} \mathcal{P} \frac{1}{1 - \frac{\omega^2}{\omega_0^2}} \quad (3.43)$$

$$\text{and } \chi''_{xx} = \frac{\pi \chi_0}{2} A_2 \delta(\omega - \omega_0) \quad (3.44)$$

$$\text{where } \chi_0 = \frac{2\sigma \mu_B^2}{\omega_0}$$

In writing eqns (3.43) and (3.44) we have used the following identity

$$\lim_{\epsilon \rightarrow 0} \frac{1}{x \pm i\epsilon} = \mathcal{P} \frac{1}{x} \mp i\pi \delta(x)$$

Here P denotes the principal value

The imaginary part χ''_{xx} of the dynamic susceptibility characterizes the absorption of energy by the spin system from the radiation field. In the approximation considered here, this absorption occurs at a frequency of the r.f. field equal to ω_0 .

Kittel³ was the first to obtain the value of the resonance frequency using the equation of motion method. He obtained the following expression

$$\omega_0 = 2\mu_B \left[\left\{ B + (N_x - N_z)M \right\} \left\{ B + (N_y - N_z)M \right\} \right]^{\frac{1}{2}} \quad (3.45)$$

The resonant frequency expression (3.41) obtained by Green function method is a generalization of Kittel's formula (3.45) and is valid at all temperatures.

From eqn (3.41) it is clear that for an unbounded medium, i.e., for a ferromagnet of infinite dimensions ω_0 reduces to

$$\omega_0 = 2\mu_B B \quad \text{Since } N_1 = 0$$

The resonance frequency, therefore, evidently depends on the shape of the specimen. It has also been established experimentally^{60,62}

3.5 Discussion

We notice that in the present approximation the ferromagnetic resonance line turns out to be delta-function

shaped. This is connected with the fact that in the present approximation the Green functions have poles on the real axis. In the higher approximation (spin wave - spin wave processes) the Green functions would have a cut along the real axis and, therefore, one would get damping. It should be pointed out that the coupling of magnons with other excitations of the system such as phonons and conduction electrons would lead to lines of finite width.

In the next section we shall consider magnon-phonon coupling. The coupling of the magnons with conduction electrons will be considered in Chapter IV.

We have considered here only the demagnetizing effects of the surface of the finite sample. The volume demagnetizing fields have not been included. Actually the dipolar interaction can give rise to volume demagnetization which produces elliptical precession instead of circular precession. The ellipticity is maximum for $\frac{\pi}{2}$ magnons, i.e., when the angle between the spin wave vector and the applied static field is 90° . Haas⁷⁶ used the dipolar Hamiltonian and calculated the renormalized spin wave energies. The results obtained by him for uniform precession mode using Callen⁷⁷ decoupling procedure are found to be identical with those of the spin wave theory and extend them through the entire temperature range.

3.6 Magnon-Phonon Coupling

It has already been pointed out in Chapter I that in

ferromagnetic insulators the dominant relaxation mechanisms are due to magnon-magnon and magnon-phonon interactions. The magnon-magnon interactions arise out of the exchange and dipolar interactions. Various workers have investigated these processes extensively in garnets. The magnon-phonon processes originate from the magnetic anisotropy, magnetostriction¹⁴ and dipolar interactions¹³. The observed magnetostriction can be accounted for by a free energy density of the form

$$f = B_1(\alpha^2 e_{xx} + \beta^2 e_{yy} + \gamma^2 e_{zz}) + B_2(\alpha\beta e_{xy} + \beta\gamma e_{yz} + \gamma\alpha e_{zx}) \quad (3.46)$$

where B_1 and B_2 are constants to be determined from the experiment. α , β and γ denote the direction cosines of the magnetization and e 's denote the elastic strain components. The strain components can be obtained by differentiating the displacement with respect to appropriate spatial coordinates. The displacement u_i (of the atom or ion at i^{th} lattice site) can be expressed in terms of phonon creation and annihilation operators $b_{\vec{K}}^{\dagger}$ and $b_{\vec{K}}$ as follows

$$u_i = \left(\frac{1}{V}\right)^{\frac{1}{2}} \sum_{\vec{K}} q_{\vec{K}} e^{i\vec{K} \cdot \vec{r}_i} \quad (3.47)$$

$$\text{where } q_{\vec{K}} = \left(\frac{\hbar}{2\rho V}\right)^{\frac{1}{2}} \sum_{s=1}^3 C_s^{-\frac{1}{2}} \xi_s \left[b_{\vec{K}}^{(s)} + b_{-\vec{K}}^{(s)\dagger} \right]$$

Here V is the volume and ρ is the density of the material, ξ_s denotes an orthogonal set of polarization vectors, C_s denote the phonon velocities ($C_1 = C_l$ is the longitudinal velocity and $C_{2,3} = C_t$ is the transverse velocity).

Using (3.46) and (3.47) the following expression for the Hamiltonian in the second quantized form can be obtained

$$H = \sum_{\substack{\vec{k}, \vec{K}, \vec{k} \\ \vec{k} = \vec{k}' + \vec{K}}} \left[E(\vec{K}) a_{\vec{k}} a_{\vec{k}'}^{\dagger} b_{\vec{K}}^{\dagger} + E^*(\vec{K}) a_{\vec{k}}^{\dagger} a_{\vec{k}'} b_{\vec{K}} \right] \quad (3.48)$$

where $E(\vec{K}) = \text{constant}(\vec{K})^{\frac{1}{2}}$, describes the coupling of magnons with phonons. $a_{\vec{k}}^{\dagger}$ and $a_{\vec{k}}$ denote respectively the creation and annihilation operators for magnons of wave vector \vec{k} . The terms in (3.48) represent two magnon - one phonon processes. The first term says that a magnon of wave vector \vec{k} and a phonon of wave vector \vec{K} annihilate to create a magnon of wave vector \vec{k} . The second term denotes the inverse process, i.e., a magnon of wave vector \vec{k} is annihilated to create another magnon and a phonon. The energy and momenta are conserved in these processes. There may be other processes such as one magnon - one phonon which have been ignored since they contribute insignificantly⁷⁸ in YIG. Kasuya et al⁷⁸ have shown that one magnon - two phonon relaxation processes are also unimportant in YIG. The magnon phonon relaxation processes are not very important in the case of metals.

3.7 Hamiltonian for the Coupled Magnon-Phonon System

Considering a system of magnons interacting with phonons the Hamiltonian for such a system can be written in the form

$$H = H_m + H_p + H_{m-p} \quad (3.49)$$



$$\text{where } H_m = \sum_k \omega_k (a_k^\dagger a_k + \frac{1}{2})$$

$$H_p = \sum_k \epsilon_k (b_K^\dagger b_K + \frac{1}{2})$$

$$\text{and } H_{m-p} = \sum_{\substack{k, k', K \\ k=k'+K}} \left[E(K) a_k a_{k'}^\dagger b_K^\dagger + E^*(K) a_k^\dagger a_{k'} b_K \right]$$

$$n_k = a_k^\dagger a_k, \quad N_K = b_K^\dagger b_K$$

Here $\omega_k = g \mu_B B + Dk^2$ is the spin wave energy and $\epsilon_k = k$, is the phonon energy D is the spin wave coupling constant

The various terms H_m , H_p and H_{m-p} in the Hamiltonian (3.49) denote respectively magnon, phonon and magnon-phonon Hamiltonians. a_k^\dagger , a_k and b_K^\dagger , b_K have already been defined. n_k and N_K represent the occupation number operators for magnons and phonons respectively $E(K)$ which has already been defined describes the strength of the coupling of these two excitations.

3.8 Magnon and Phonon Green Functions

We define single magnon and phonon Green functions as follows

$$\begin{aligned} G_{qq}^m(t) &= \langle\langle a_q(t), a_q^\dagger(0) \rangle\rangle \\ G_{qq}^p(t) &= \langle\langle b_q(t), b_q^\dagger(0) \rangle\rangle \end{aligned} \quad (3.50)$$

Using eqn. (3.3), we write down the equations of motion for their Fourier transforms

$$(\omega - \omega_q) G_{qq}^m(\omega) = \frac{1}{2\pi} + \sum_K E(K) F_{q+K,K,q}^+(\omega) + \sum_K E^*(K) F_{q-K,K,q}(\omega) \quad (3.51)$$

$$\text{and } (\omega - \varepsilon_q) G_{qq}^p(\omega) = \frac{1}{2\pi} + E(q) \sum_k T_{k+q,k,q}(\omega)$$

where we have introduced the following mixed Green functions

$$\begin{aligned} F_{q+K,K,q}^+(\omega) &= \ll a_{q+K} b_K^+, a_q^+ \gg_\omega \\ F_{q-K,K,q}(\omega) &= \ll a_{q+K} b_K^-, a_q^+ \gg_\omega \end{aligned} \quad (3.52)$$

and

$$T_{k+q,k,q}(\omega) = \ll a_{k+q} a_k^+, b_q^+ \gg_\omega$$

We construct the equations of motion for these mixed Green functions and get

$$\begin{aligned} (\omega - \omega_{q+K} + \varepsilon_K) F_{q+K,K,q}^+ &= \sum_k E(k) \ll a_{q+K+k} b_K^+ b_k^+, a_q^+ \gg_\omega \\ &+ \sum_k E^*(k) \ll a_{q+K-k} b_K^+ b_k^-, a_q^+ \gg_\omega \\ &- E^*(K) \sum_k \ll a_{k+K}^+ a_k a_{q+K}, a_q^+ \gg_\omega \end{aligned} \quad (3.53a)$$

$$\begin{aligned} (\omega - \omega_{q-K} - \varepsilon_K) F_{q-K,K,q} &= \sum_k E(k) \ll a_{q-K+k} b_K^- b_k^+, a_q^+ \gg_\omega \\ &+ E(K) \sum_k \ll a_k^+ a_{k+K} a_{q-K}, a_q^+ \gg_\omega \\ &+ \sum_k E^*(k) \ll a_{q-K+k} b_K^- b_k^-, a_q^+ \gg_\omega \end{aligned} \quad (3.53b)$$

and

$$\begin{aligned} (\omega - \omega_{k+q} + \omega_k) T_{k+q,k,q}(\omega) &= - \sum_{k_1} E(k_1) \ll a_{k+q} a_{k-k_1}^+ b_{k_1}^+, b_q^+ \gg_\omega \\ &+ \sum_{k_1} E(k_1) \ll a_{k+q+k_1} a_k^+ b_{k_1}^+, b_q^+ \gg_\omega \\ &+ \sum_{k_1} E^*(k_1) \ll a_k^+ a_{k+q-k_1} b_{k_1}^-, b_q^+ \gg_\omega \\ &- \sum_{k_1} E^*(k_1) \ll a_{k+k_1}^+ a_{k+q} b_{k_1}^-, b_q^+ \gg_\omega \end{aligned} \quad (3.53c)$$

These equations (3.53) do not contain an inhomogeneous part as $K \neq 0$, i.e., $E(K) = 0$ for $K = 0$.

We notice that eqns. (3.53) contain still higher order Green functions for which we should, in turn, construct equations of motion and continue the process further. We do not wish to attempt this here and decouple the higher order Green functions as follows

$$\begin{aligned} \langle\langle a_{q+K+k} b_K^+ b_k^+, a_q^+ \rangle\rangle_\omega &\approx \langle b_K^+ b_k^+ \rangle G_{q+K+k,q}^m(\omega) = 0 \\ \langle\langle a_{q+K-k} b_K^+ b_k^+, a_q^+ \rangle\rangle_\omega &\approx \bar{n}_K \delta_{Kk} G_{qq}^m(\omega) \end{aligned} \quad (3.54a)$$

$$\begin{aligned} \langle\langle a_{k+K}^+ a_k a_{q+K}, a_q^+ \rangle\rangle_\omega &\approx \bar{n}_{q+K} \delta_{qk} G_{qq}^m(\omega) \\ \langle\langle a_{q-K} b_K b_k^+, a_q^+ \rangle\rangle_\omega &\approx (1 + \bar{n}_K) \delta_{Kk} G_{qq}^m(\omega) \\ \langle\langle a_k^+ a_{k+K} a_{q-K}, a_q^+ \rangle\rangle_\omega &\approx \bar{n}_{q-K} \delta_{k,q-K} G_{qq}^m(\omega) \end{aligned} \quad (3.54b)$$

$$\begin{aligned} \langle\langle a_{q-K-k} b_K b_k^+, a_q^+ \rangle\rangle_\omega &\approx \langle b_K b_k^+ \rangle G_{q-K-k,q}^m(\omega) = 0 \\ \langle\langle a_{k+q} a_{k-k_1}^+ b_{k_1}^+, b_q^+ \rangle\rangle_\omega &\approx \langle a_{k+q} a_{k-k_1}^+ \rangle \langle\langle b_{k_1}^+, b_q^+ \rangle\rangle = 0 \\ \langle\langle a_{k+q+k_1} a_k^+ b_{k_1}^+, b_q^+ \rangle\rangle_\omega &\approx \langle a_{k+q+k_1} a_k^+ \rangle \langle\langle b_{k_1}^+, b_q^+ \rangle\rangle = 0 \quad (3.54c) \\ \langle\langle a_k^+ a_{k+q-k_1} b_{k_1}, b_q^+ \rangle\rangle_\omega &\approx \bar{n}_k \delta_{qk_1} G_{qq}^p(\omega) \end{aligned}$$

$$\text{and } \langle\langle a_{k+k_1}^+ a_{k+q} b_{k_1}, b_q^+ \rangle\rangle_\omega \approx \bar{n}_{k+q} \delta_{qk_1} G_{qq}^p(\omega)$$

$$\text{where } \bar{n}_k = \langle a_k^+ a_k \rangle \quad \text{and} \quad \bar{n}_K = \langle b_K^+ b_K \rangle$$

In the above equations the correlations between two creation and two annihilation operators have been neglected

With these approximations (3.54), eqns (3.51) and (3.53) yield

$$G_{qq}^m(\omega) = \frac{1}{2\pi [\omega - \omega_q - M_q^m(\omega)]} \quad (3.55a)$$

$$\text{and } G_{qq}^p(\omega) = \frac{1}{2\pi [\omega - \varepsilon_q - M_q^p(\omega)]} \quad (3.55b)$$

where

$$M_q^m(\omega) = \sum_K |E(K)|^2 \left[\frac{\bar{N}_K - \bar{n}_{q+K}}{\omega - \omega_{q+K} + \varepsilon_K} + \frac{1 + \bar{N}_K + \bar{n}_{q-K}}{\omega - \omega_{q-K} - \varepsilon_K} \right] \quad (3.56a)$$

$$\text{and } M_q^p(\omega) = |E(q)|^2 \sum_K \frac{\bar{n}_K - \bar{n}_{K+q}}{\omega - \omega_{K+q} + \omega_K} \quad (3.56b)$$

The poles of Green functions $G_{qq}^m(\omega)$ and $G_{qq}^p(\omega)$ respectively are given by

$$\omega - \omega_q - M_q^m(\omega) = 0 \quad (3.57a)$$

$$\text{and } \omega - \varepsilon_q - M_q^p(\omega) = 0 \quad (3.57b)$$

Using the identity

$$\lim_{\varepsilon \rightarrow 0} \frac{1}{x \pm i\varepsilon} = \frac{P}{x} \mp i\pi\delta(x)$$

We write down for the real and imaginary parts of

$M_q^m(\omega)$ and $M_q^p(\omega)$ respectively

$$R_q^m(\omega) = \text{Re } M_q^m(\omega) = P \sum_K |E(K)|^2 \left[\frac{\bar{N}_K - \bar{n}_{q+K}}{\omega - \omega_{q+K} + \varepsilon_K} + \frac{1 + \bar{N}_K + \bar{n}_{q-K}}{\omega - \omega_{q-K} - \varepsilon_K} \right] \quad (3.58a)$$

$$\text{Im } M_q^m(\omega) = \gamma_q^m(\omega) = \pi \sum_K |E(K)|^2 \left[(\bar{N}_K - \bar{n}_{q+K}) \delta(\omega - \omega_{q+K} + \varepsilon_K) + (1 + \bar{N}_K + \bar{n}_{q-K}) \delta(\omega - \omega_{q-K} - \varepsilon_K) \right] \quad (3.58b)$$

$$R_q^P(\omega) = \text{Re } M_q^P(\omega) = P |E(q)|^2 \sum_K \frac{\bar{n}_K - \bar{n}_{K+q}}{(\omega - \omega_{K+q} + \omega_K)} \quad (3.59a)$$

and

$$\gamma_q^P(\omega) = \text{Im } M_q^P(\omega) = |E(q)|^2 \sum_K (\bar{n}_K - \bar{n}_{K+q}) \delta(\omega - \omega_{K+q} + \omega_K) \quad (3.59b)$$

where P indicates that the principal value of the corresponding integral should be taken

The functions $\gamma_q^m(\omega)$ and $\gamma_q^P(\omega)$ play the role of damping for magnon and phonon excitations. $R_q^m(\omega)$ and $R_q^P(\omega)$ denote the corresponding energy shifts for them

Using eqns. (3.6), (3.55), (3.58) and (3.59) we can obtain for magnon and phonon distribution functions

$$\bar{n}_q = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\gamma_q^m(\omega) (e^{\beta\omega} - 1)^{-1} d\omega}{\left[\omega - \omega_q - M_q^m(\omega) \right]^2 + \left[\gamma_q^m(\omega) \right]^2} \quad (3.60a)$$

$$\text{and } \bar{N}_q = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\gamma_q^P(\omega) (e^{\beta\omega} - 1)^{-1} d\omega}{\left[\omega - \varepsilon_q - M_q^P(\omega) \right]^2 + \left[\gamma_q^P(\omega) \right]^2} \quad (3.60b)$$

If $\gamma_q^m(\omega)$, $\gamma_q^P(\omega)$ and $R_q^m(\omega)$ and $R_q^P(\omega)$ are slowly varying functions of ω and if $\gamma_q^m(\omega)$ and $\gamma_q^P(\omega)$ are smaller than $R_q^m(\omega)$ and $R_q^P(\omega)$ respectively then eqns (3.60) assume the following form

$$\bar{n}_q = \frac{1}{e^{\beta\omega_q} - 1} \quad (3.61a)$$

$$\text{and} \quad \bar{N}_q = \frac{1}{e^{\beta \tilde{\epsilon}_q - 1}} \quad (3.61b)$$

where $\tilde{\omega}_q$ and $\tilde{\epsilon}_q$ are determined by

$$\tilde{\omega}_q - \omega_q - M_q^m(\tilde{\omega}_q) = 0 \quad (3.62a)$$

$$\text{and} \quad \tilde{\epsilon}_q - \epsilon_q - M_q^p(\tilde{\epsilon}_q) = 0 \quad (3.62b)$$

We notice that \bar{n}_q and \bar{N}_q are Bose-Einstein distribution functions for magnons and phonons which are "smeared out" because of magnon-phonon interaction. The widths of the "smearing out" regions are of the order of the magnitude of the damping $\tilde{\gamma}_q^m(\tilde{\omega}_q)$ and $\tilde{\gamma}_q^p(\tilde{\epsilon}_q)$ respectively.

The elementary magnon and phonon excitations are given by^{79,80}

$$\tilde{\omega}_q = \omega_q + P \sum_K |E(K)|^2 \left[\frac{\bar{N}_K - \bar{n}_{q+K}}{(\tilde{\omega}_q - \omega_{q+K} + \epsilon_K)} + \frac{1 + \bar{N}_K + \bar{n}_{q-K}}{(\tilde{\omega}_q - \omega_{q-K} - \epsilon_K)} \right] \quad (3.63a)$$

$$\text{and} \quad \tilde{\epsilon}_q = \epsilon_q + |E(q)|^2 P \sum_k \frac{(\bar{n}_k - \bar{n}_{k+q})}{(\tilde{\epsilon}_q - \omega_{k+q} + \omega_k)} \quad (3.63b)$$

We observe that the magnon and phonon energies are renormalized as a result of the magnon-phonon interaction

CHAPTER - IV

FERROMAGNETIC RELAXATION IN METALS "s-d EXCHANGE" INTERACTION4.1 Spin Waves in Metals

In the Heisenberg model of a ferromagnet besides the spin aligned state constituting the ground state, there exist spin wave states close to the ground state. For metals where the electrons are assumed to be itinerant, it was not clear for quite sometime whether spin waves could exist in them. The existence of spin waves in both ferromagnetic metals and insulators has been established experimentally⁸¹. Herring and Kittel⁸² who proposed that spin^{waves}/did exist in metals showed that the magnetization of a metal responded to a magnetic field with a sinusoidal spatial dependence in such a way as to indicate the possibility of spin wave like excitations.

In an itinerant electron model, the electrons are divided between 'up' and 'down' bands. These 'up' and 'down' spin bands are displaced relative to each other due to an exchange interaction which acts just like an internal magnetic field⁸³. Consequently, the wave vector for the electron in the highest filled state will be different for 'up' and 'down' spins. An electron in the 'down' band can be excited to 'up' band without any change in the wave vector of the electron, provided it is given sufficient energy, while an electron from 'up' band can only make a transition to 'down' if its wave vector changes by at least Δk_F , the

horizontal spacing between the bands

These transitions are described as single particle excitations as we reverse the spin of a particular electron. Using all the electrons, a collective excitation can be obtained. This is done for the 'down' to 'up' transition in which, as stated earlier, the wave vector change may tend to zero and it is this excitation which is assumed to be a spin wave in the itinerant electron model. The wave vector of this spin wave will be equal to q , the change in wave vector of the electrons as they go from one band to another

We discuss spin waves in an electron gas which is assumed to have a net polarization.

The Hamiltonian of the system can be written in the form⁸⁴

$$H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} C_{\mathbf{k}\uparrow}^{\dagger} C_{\mathbf{k}\uparrow} + \frac{1}{2} \sum_{\mathbf{q}, \mathbf{k}, \mathbf{k}'} \sum_{\sigma, \sigma'} V(\mathbf{q}) C_{\mathbf{k}+\mathbf{q}, \sigma}^{\dagger} C_{\mathbf{k}', -\mathbf{q}, \sigma'}^{\dagger} C_{\mathbf{k}'\sigma'} C_{\mathbf{k}\sigma} \quad (4.1)$$

where $C_{\mathbf{k}\sigma}^{\dagger}$ and $C_{\mathbf{k}\sigma}$ denote creation and annihilation operators for Bloch waves of wave vector \vec{k} and spin σ . $V(\mathbf{q})$ is the Fourier transform of the Coulomb potential

We define a quantity $S_{\mathbf{k}}^{-}(\mathbf{q})$

$$S_{\mathbf{k}}^{-}(\mathbf{q}) = C_{\mathbf{k}+\mathbf{q}\uparrow}^{\dagger} C_{\mathbf{k}\downarrow} \quad (4.2)$$

which describes an excitation in which an electron with down spin in the state \mathbf{k} is removed into a state $\mathbf{k}+\mathbf{q}$ with spin up. The equation of motion of this quantity can be written as

$$i \frac{dS_{\mathbf{k}}^{-}(\mathbf{q})}{dt} = [S_{\mathbf{k}}^{-}(\mathbf{q}), H] \quad (4.3)$$

The commutator of $S_k^-(q)$ with the Hamiltonian (4.1) can be evaluated to yield

$$i \frac{dS_k^-(q)}{dt} = (\epsilon_k - \epsilon_{k+q}) S_k^-(q) + \sum_{k_1 k_2} v(k_2) \left[C_{k_1+k_2, \sigma}^+ C_{k+q \uparrow}^+ \times \right. \\ \left. C_{k+k_2, \downarrow} C_{k_1, \sigma} - C_{k+q+k_2, \uparrow}^+ C_{k_1-k_2, \sigma}^+ C_{k_1, \sigma} C_{k, \downarrow} \right] \quad (4.4)$$

We apply random phase approximation (RPA) to the right hand side of eqn. (4.4) and retain only such terms in the four operator products as contain pairs of operators C_k^+ & $C_{k, \sigma}$ and replace them by $n_{k, \sigma}$, where $n_{k, \sigma}$ is the mean value of the number operator $C_k^+ C_{k, \sigma}$ in the ground state. The equation of motion (4.4) now reduces to

$$i \frac{dS_k^-(q)}{dt} = (\epsilon_k - \epsilon_{k+q}) S_k^-(q) + \sum_{k_1} v(k_1) \left[\bar{n}_{k+k_1+q \uparrow} - \bar{n}_{k+k_1 \downarrow} \right] S_k^-(q) \\ + (\bar{n}_{k \downarrow} - \bar{n}_{k+q \uparrow}) \sum_{k_1} v(k_1) S_{k+k_1}^-(q) \quad (4.4')$$

Introducing the renormalized energies $\omega_{k, \sigma}$ through

$$\omega_{k \downarrow} = \epsilon_k - \sum_{k_1} v(k_1) \bar{n}_{k+k_1 \downarrow} \\ \omega_{k \uparrow} = \epsilon_k - \sum_{k_1} v(k_1) \bar{n}_{k+k_1 \uparrow} \quad (4.5)$$

and assuming that the ground state is ferromagnetic so that the numbers of the up and down electrons are unequal and so are the energies $\omega_{k \uparrow}$ and $\omega_{k \downarrow}$, we have

$$(i \frac{d}{dt} + \omega_{k+q \uparrow} - \omega_{k \downarrow}) S_k^-(q) + (\bar{n}_{k+q \uparrow} - \bar{n}_{k \downarrow}) \sum_{k_1} v(k_1) S_{k+k_1}^-(q) = 0 \quad (4.6)$$

If the states of the system are assumed to be of the form

$$|\Psi\rangle = \sum_{\mathbf{k}} A_{\mathbf{k}} S_{\mathbf{k}}^{-}(\mathbf{q}) |0\rangle$$

with energy ω , then the RPA equation for the coefficients $A_{\mathbf{k}}$ can be written as follows

$$(\omega_{\mathbf{k}+\mathbf{q}\uparrow} - \omega_{\mathbf{k}\downarrow} - \omega) A_{\mathbf{k}} + \sum_{\mathbf{k}_1} V(\mathbf{k}_1) [\tilde{n}_{\mathbf{k}-\mathbf{k}_1+\mathbf{q}\uparrow} - \tilde{n}_{\mathbf{k}-\mathbf{k}_1\downarrow}] A_{\mathbf{k}-\mathbf{k}_1} = 0 \quad (4.7)$$

It is obvious that such states, if they exist, will resemble spin waves since they reverse one unit of spin

As mentioned earlier, $V(\mathbf{k}_1)$ is the Fourier transform of the Coulomb interaction. However, if correlation effects are taken into account the screened Coulomb interaction will be more appropriate. Replacing its Fourier transform by a constant V ($V > 0$) we obtain

$$1 + V \sum_{\mathbf{k}} \frac{(\tilde{n}_{\mathbf{k}+\mathbf{q}\uparrow} - \tilde{n}_{\mathbf{k}\downarrow})}{\omega_{\mathbf{k}+\mathbf{q}\uparrow} - \omega_{\mathbf{k}\downarrow} - \omega} = 0 \quad (4.8)$$

The relations (4.5) now reduce to

$$\omega_{\mathbf{k}\downarrow} = \epsilon_{\mathbf{k}} - N_{\downarrow} V \quad (4.9)$$

and
$$\omega_{\mathbf{k}\uparrow} = \epsilon_{\mathbf{k}} - N_{\uparrow} V$$

where N_{\downarrow} and N_{\uparrow} denote total number of electrons with down and up spin respectively. We assume that $N_{\uparrow} > N_{\downarrow}$ and consequently $\omega_{\mathbf{k}\uparrow} < \omega_{\mathbf{k}\downarrow}$.

We notice that the second terms in the right hand side of the eqns. (4.5) and (4.9) denote the exchange energy of

the electron. It appears that the Coulomb interaction is responsible for displacing the spin bands for up and down electrons. However, some authors are of the opinion that this displacement of bands is caused by the exchange interaction.

For vanishingly small value of q the eqn (4.8) takes the following form

$$1 + V \sum_{\substack{\omega_{k\downarrow} > \epsilon_F \\ \omega_{k+q\uparrow} < \epsilon_F}} \frac{1}{(N_{\downarrow} - N_{\uparrow})V - \omega} = 0 \quad (4.10)$$

Using the eqns. (4.9) and (4.10) we obtain

$$1 - V \sum_{\substack{\epsilon_{k+q} < \epsilon_F + N_{\uparrow}V \\ \epsilon_k > \epsilon_F + N_{\downarrow}V}} \frac{1}{V(N_{\uparrow} - N_{\downarrow}) - \epsilon_{k+q} + \epsilon_k + \omega} \quad (4.11)$$

or

$$1 = \frac{1}{(N_{\uparrow} - N_{\downarrow})} \sum_{\substack{\epsilon_{k+q} < \epsilon_F + N_{\uparrow}V \\ \epsilon_k > \epsilon_F + N_{\downarrow}V}} \frac{1}{V(N_{\uparrow} - N_{\downarrow})} + \frac{1}{V(N_{\uparrow} - N_{\downarrow})^2} \sum_{\substack{\epsilon_F + N_{\downarrow}V < \epsilon_k \\ < \epsilon_F + N_{\uparrow}V}} (\epsilon_{k+q} + \epsilon_k - \omega)$$

We now have

$$\omega = \frac{1}{(N_{\uparrow} - N_{\downarrow})} \sum \left[(q \cdot \nabla_k) \epsilon_k + \frac{1}{2} (q \cdot \nabla_k)^2 \epsilon_k + \dots \right] \quad (4.12)$$

If $\epsilon_k = \epsilon_{-k}$ the first term of (4.12) vanishes and

we get

$$\omega = \frac{1}{2} \langle (q \cdot \nabla_k)^2 \epsilon_k \rangle \quad (4.13)$$

where the average $\langle \dots \rangle$ is taken over the region between the

two Fermi surfaces

This establishes that there are low lying excitations of the spin wave type . Thus we have the same spectrum as spin waves for the localized spin model. It should be pointed out that in this calculation $V(q)$ has been replaced by a constant V greater than zero, which means that $V(r)$ in real space is a delta function This is not unreasonable in that the interactions are strongly screened by conduction electrons. While this particular type of simplification is not essential to bring out the general nature of the excitations, it does show that some type of localization is brought into the model

Rajagopal et al⁴⁰ have studied single particle and collective excitations in an itinerant electron model More recently, Sokoloff⁸⁵ has also investigated polar spin waves in ferromagnetic metals.

4.2 "s-d Exchange" Interaction

The spin wave system can also relax through the interaction with the conduction electrons This interaction can be brought about by the "s-d exchange" interaction. This is regarded as the most predominant interaction in ferromagnetic transition metals. A number of workers⁴¹⁻⁴⁴ have studied this interaction in detail In the present work, this interaction has been studied to explain relaxation in these metals. It has already been pointed out in Chapter I that

the exchange interaction between 3d-electrons is mediated by conduction s electrons. Actually, owing to the "s-d exchange" interaction between conduction electrons and the localized 3d spin at i^{th} lattice site, the conduction electron spin system is polarized and consequently the spin polarization of conduction electrons appears around the i^{th} site. Usually most of the induced spin polarization is localized at i^{th} site and contributes to self energy. A part of the induced spin polarization may extend to other lattice sites j and induce an effective exchange field on the 3d electron spin localized at j^{th} site through "s-d exchange" interaction.

4.3 Hamiltonian

The Hamiltonian for the 3d-localized ferromagnetic electrons exchange coupled with conduction s electrons is of the form

$$H = H_z + H_c + H_{ex} + H_{s-d} \quad (4.14)$$

$$\text{where } H_z = -g\mu_B B \sum_n S_n^z \quad (4.14a)$$

$$H_c = \sum_{k\sigma} \epsilon_k' C_{k\sigma}^+ C_{k\sigma} \quad (4.14b)$$

$$H_{ex} = - \sum_{n>m} J(\vec{m} - \vec{n}) \vec{S}_m \cdot \vec{S}_n \quad (4.14c)$$

$$\text{and } H_{s-d} = - \sum_{mn} J_{s-d}(\vec{r}_m - \vec{r}_n) \vec{s}_m \cdot \vec{S}_n \quad (4.14d)$$

The various symbols have their usual meaning. The three terms on the right hand side of eqn. (4.14) denote

respectively Zeeman, conduction electron, exchange and "s-d exchange" energies. In terms of the creation and annihilation operators $c_{k\sigma}^\dagger$ and $c_{k\sigma}$ ($\sigma = \pm \frac{1}{2}$), the "s-d exchange" Hamiltonian (4.14d) can be written in the form

$$H_{s-d} = - \frac{1}{N} \sum_{nkk'} J_{s-d}(kk') e^{i(k'-k) \cdot R_n} \left[c_{k-}^+ c_k + S_n^- + c_{k+}^+ c_{k'} - S_n^+ + (c_{k-}^+ c_{k'} - c_{k+}^+ c_{k'}) S_n^z \right] \quad (4.14d)$$

where $S_n^\pm = S_n^x \pm i S_n^y$, $J_{s-d}(kk')$ is the s-d exchange integral given by

$$J_{s-d}(kk') = \iint \psi_k^*(r) \phi^*(r') \frac{e^2}{|r_{12}|} \psi_{k'}(r') \phi(r) d^3r d^3r'$$

where $\phi(r)$ is the atomic function for d-electron and ψ_k is the Bloch electron wave function.

Introducing Hoshen-Primakoff transformation⁸⁶ and using spin wave approximation, i.e., $S_n^- = b_n^+$, $S_n^+ = b_n$ where b_n^+ and b_n create and destroy respectively spin deviation at the lattice site n , the Hamiltonian (4.14) can be expressed as follows

$$H = \sum_{k\sigma} \xi_k^\sigma c_{k\sigma}^\dagger c_{k\sigma} + \sum_q \varepsilon_q b_q^+ b_q - \left(\frac{2S}{N}\right)^{\frac{1}{2}} \sum_{kk'q} J_{s-d}(kk') \times \\ \delta(k' - k + q) \left[c_{k+}^+ c_{k'} b_q + c_{k-}^+ c_{k+} b_q^+ \right] + \frac{1}{N} \sum_{kk'qq'} J_{s-d}(kk') \times \\ \delta(k' - k + q' - q) \left[c_{k-}^+ c_{k'} - c_{k+}^+ c_{k'} \right] b_q^+ b_{q'} \quad (4.15)$$

where b_q^+ and b_q are creation and annihilation operators for spin-waves of wave vector q , and

$$\varepsilon_q^\sigma = \varepsilon_q^0 - s J_{s-d}(qq) + 2 \sigma \mu_B B, \quad \varepsilon_q^0 = \frac{q^2}{2m}, \quad \varepsilon_q = Dq^2 + g\mu_B B$$

The 3rd and 4th terms represent the coupling of magnons with conduction electrons. The 3rd term denotes the scattering of an s electron from one Bloch state to the other including a spin flip, and simultaneous creation and destruction of a spin wave. The 4th term represents s electron scattering without spin flip and spin wave scattering from state q to q'.

4.4 Magnon Green Functions

We define a single magnon Green function

$$G_{qq_1}(t) = \ll b_q(t), b_{q_1}^+(0) \gg = -i\theta(t) \langle [b_q, b_{q_1}^+] \rangle \quad (4.16)$$

Here, as has been pointed earlier, $\langle - \rangle$ denotes averaging over a grand canonical ensemble at a given temperature, $\theta(t) = 1$ if $t > 0$, and zero otherwise. Here all the operators are in Heisenberg representation.

The time derivative of $G_{qq_1}(t)$ is given by

$$\begin{aligned} i \frac{dG_{qq_1}(t)}{dt} &= \delta(t) \delta_{qq_1} + \varepsilon_q G_{qq_1}(t) - \left(\frac{2S}{N}\right)^{\frac{1}{2}} \sum_k J_{s-d}(k, k-q) \times \\ &\quad \ll c_{k-q}^+ c_{k+}, b_{q_1}^+ \gg + \frac{1}{N} \sum_{kk_1} J_{s-d}(kk_1) \times \\ &\quad \ll (c_{k-}^+ c_{k_1-} - c_{k+}^+ c_{k_1+}) b_{k+q-k_1}, b_{q_1}^+ \gg \quad (4.17) \end{aligned}$$

Here we notice appearance of higher order Green functions. We use the following decoupling approximations to decouple them

$$\begin{aligned}
\langle\langle c_{k-}^+ c_{k_1-} b_{k+q-k_1}, b_{q_1}^+ \rangle\rangle &\approx \bar{n}_{k-} \delta_{kk_1} G_{qq_1}(-) \\
\langle\langle c_{k+}^+ c_{k_1+} b_{k+q-k_1}, b_{q_1}^+ \rangle\rangle &\approx \bar{n}_{k+} \delta_{kk_1} G_{qq_1}(+)
\end{aligned} \quad (4.18)$$

where $\bar{n}_{k-} = \langle c_{k-}^+ c_{k-} \rangle$, $\bar{n}_{k+} = \langle c_{k+}^+ c_{k+} \rangle$

Fourier transformation of (4.17) with equation (4.18)

yields

$$\begin{aligned}
\left[\omega - \epsilon_q - \frac{1}{N} \sum_k J_{s-d}(k, k) (\bar{n}_{k-} - \bar{n}_{k+}) \right] G_{qq_1}(\omega) \\
= \frac{1}{2\pi} - \left(\frac{2S}{N} \right)^{\frac{1}{2}} \sum_k J_{s-d}(k, k-q) F_{k-q, k, q_1}(\omega) \quad (4.19)
\end{aligned}$$

where we have substituted

$$G_{qq_1}(t) = N^{-1} \int d\omega G_{qq_1}(\omega) e^{i\omega t} \quad (4.20)$$

and $F_{k-q, k, q_1}(\omega) = \langle\langle c_{k-q-}^+ c_{k+}, b_{q_1}^+ \rangle\rangle_{\omega}$

To obtain $G_{qq_1}(\omega)$ from eqn (4.19), we should write down the equation of motion for $F_{k-q, k, q_1}(t)$. Its time derivative is given by

$$\begin{aligned}
\frac{dF_{k-q, k, q_1}(t)}{dt} &= (\epsilon_k^+ - \epsilon_{k-q}^-) F_{k-q, q, q_1}(t) - \left(\frac{2S}{N} \right)^{\frac{1}{2}} \times \\
&\quad \left[\sum_p J_{s-d}(k, k-p) \langle\langle c_{k-q-}^+ c_{k-p-} b_p, b_{q_1}^+ \rangle\rangle \right. \\
&\quad \left. - \sum_p J_{s-d}(p, k-q) \langle\langle c_{p+}^+ c_{k+} b_{p+q-k}, b_{q_1}^+ \rangle\rangle \right] \\
&\quad - \frac{1}{N} \left[\sum_{p_0 p'} J_{s-d}(p_0, k-q) \langle\langle c_{p_0-}^+ c_{k+} b_{p'}^+ b_{p_0+p'-k+q}, b_{q_1}^+ \rangle\rangle \right. \\
&\quad \left. + \sum_{pp_1} J_{s-d}(k, p) \langle\langle c_{k-q-}^+ c_{p+} b_{p_1}^+ b_{k+p_1-p}, b_{q_1}^+ \rangle\rangle \right] \quad (4.21)
\end{aligned}$$

These equations contain several higher order mixed Green functions which are decoupled in the following way

$$\langle\langle c_{p+}^+ c_{k+} b_{p+q-k} b_{q_1}^+ \rangle\rangle \simeq \bar{n}_{k+} \sum_{pk} G_{qq_1}(t)$$

$$\langle\langle c_{p_0}^+ c_{k+} b_{p'}^+ b_{p_0+p'-k+q} b_{q_1}^+ \rangle\rangle \simeq \bar{n}_p F_{k-q,k,q_1}(t) \delta_{p,k-q}$$

$$\langle\langle c_{k-q-}^+ c_{k-p-} b_p b_{q_1}^+ \rangle\rangle \simeq \bar{n}_{k-q-} \sum_{qp} G_{qq_1}(t)$$

$$\text{and } \langle\langle c_{k-q-}^+ c_{p+} b_{p_1}^+ b_{k+p_1-p} b_{q_1}^+ \rangle\rangle \simeq \bar{n}_{p_1} F_{k-q,k,q_1}(t) \delta_{kp} \quad (4.22)$$

where $\bar{n}_p = \langle b_p^+ b_p \rangle$, is the average number of spin waves.

Taking the Fourier transform of eqn. (4.21) and using eqns (4.18) and (4.22) we get

$$F_{k-q,k,q_1}(\omega) = -\left(\frac{2S}{N}\right)^{\frac{1}{2}} \frac{J_{s-d}(k,k-q) (\bar{n}_{k-q-} - \bar{n}_{k+})}{\omega - \varepsilon_k^+ + \varepsilon_{k-q}^- + \frac{2}{N} \sum_p J_{s-d} \bar{n}_p} G_{qq_1}(\omega) \quad (4.23)$$

$$\text{Here } \tilde{J}_{s-d} = J_{s-d}(k-q, k-q) + J_{s-d}(k,k)$$

From eqns. (4.19) and (4.23) we obtain

$$G_{qq_1}(\omega) = \frac{\mathcal{G}_{qq_1} / 2\pi}{\omega - \varepsilon_q - \Delta - \gamma_q(\omega)} \quad (4.24)$$

where we have put

$$\Delta = \frac{1}{N} \sum_k J_{s-d}(k,k) (\bar{n}_{k-} - \bar{n}_{k+}) \quad (4.25a)$$

$$\text{and } \gamma_q(\omega) = \frac{2S}{N} \sum_k \frac{J_{s-d}^2(k,k-q) (\bar{n}_{k-q-} - \bar{n}_{k+})}{\omega - \varepsilon_k^+ + \varepsilon_{k-q}^- + \frac{2}{N} \sum_p J_{s-d} \bar{n}_p} \quad (4.25b)$$

Separating the real and imaginary parts of $\int_q(\omega)$ we get

$$\text{Re } \int_q(\omega) = \frac{2S}{N} \sum_k \frac{J_{s-d}^2(k, k-q) (\bar{n}_{k-q-} - \bar{n}_{k+})}{(\omega - \varepsilon_k^+ + \varepsilon_{k-q}^- + \frac{2}{N} \sum_p \bar{n}_p)} \quad (4.26a)$$

$$\begin{aligned} \chi_q(\omega) = \text{Im } \int_q(\omega) &= \frac{2S}{N} \pi \sum_k J_{s-d}^2(k, k-q) (\bar{n}_{k-q-} - \bar{n}_{k+}) \times \\ &\quad \delta(\omega - \varepsilon_k^+ + \varepsilon_{k-q}^- + \frac{2}{N} \sum_p \bar{n}_p) \end{aligned} \quad (4.26b)$$

Here \bar{n}_p denotes the superposition of elementary Bose excitations with resonant intensity and damping $\chi_q(\omega)$, and is given by

$$\bar{n}_p = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{\chi_q(\omega) d\omega}{\left[\omega - \varepsilon_q - \frac{2}{N} \sum_p \bar{n}_p \right]^2 + \chi_q^2(\omega)} \quad (4.27)$$

We know that the response of a ferromagnet in a static magnetic field and subjected to a radiation field will be given by the magnetic susceptibility tensor defined as follows

$$\chi_{ij} = \ll \hat{M}_i(t), \hat{M}_j(t) \gg_{\omega} \quad (4.28)$$

where $\hat{M}(t)$ is the magnetization at the time t . For the present case of a ferromagnetic metal it is obvious that

$$\hat{M} = g_d \mu_B \hat{S}_d + g_s \mu_B \hat{S}_s \quad (4.29)$$

where \hat{S}_d and \hat{S}_s are the total spin operators for d and s -electrons respectively. Using (4.28) and (4.29) we have

$$\begin{aligned} \langle\langle M_y, M_y \rangle\rangle = \mu_B^2 \left[g_d^2 \langle\langle S_d^y, S_d^y \rangle\rangle + g_s g_d \langle\langle S_s^y, S_d^y \rangle\rangle \right. \\ \left. + g_s g_d \langle\langle S_d^y, S_s^y \rangle\rangle + g_s^2 \langle\langle S_s^y, S_s^y \rangle\rangle \right] \quad (4.30) \end{aligned}$$

The magnetization of s-electrons is small compared with the magnetization of d-electrons. The latter is always appreciably larger and specially at small values of the ratio J_{s-d}/ϵ_F (ϵ_F being the Fermi energy), the magnetization of the s-electrons can, therefore, be neglected. For the present case, the resonant properties of the system will be described by the Green function $\langle\langle S_d^y, S_d^y \rangle\rangle$.

In terms of the spin wave creation and annihilation operators χ_{yy} becomes

$$\chi_{yy} = \mu_B^2 g^2 \frac{SN}{2} \left\{ \langle\langle b_o, b_o^+ \rangle\rangle + \langle\langle b_o^+, b_o \rangle\rangle \right\} \quad (4.31)$$

From the above expression we notice that at resonance the spin waves of $k = 0$ should be excited in the framework of the spin wave theory. It is called uniform precession mode ferromagnetic resonance. χ_{yy} is obtained using eqn. (4.24) for $q = 0$ as follows

$$\begin{aligned} \chi_{yy} = \mu_B^2 g^2 \frac{SN}{4\pi} \left\{ \frac{\omega - \tilde{\epsilon}_o}{(\omega - \tilde{\epsilon}_o)^2 + \gamma_o^2(\omega)} + \frac{\omega + \tilde{\epsilon}_o}{(\omega + \tilde{\epsilon}_o)^2 + \gamma_o^2(\omega)} \right\} \\ + i \frac{\mu_B^2}{4\pi} g^2 SN \left\{ \frac{\gamma_o(\omega)}{(\omega - \epsilon_o)^2 + \gamma_o^2(\omega)} - \frac{\gamma_o(\omega)}{(\omega + \epsilon_o)^2 + \gamma_o^2(\omega)} \right\} \quad (4.32) \end{aligned}$$

Here $\tilde{\epsilon}_o = \epsilon_o + \Delta + \text{Re } \rho_o(\omega)$

$$\text{and } \chi_0(\omega) = \frac{2S}{N} \sum_k J_{s-d}^2(k, k) (\bar{n}_{k-} - \bar{n}_{k+}) \delta(\omega - \epsilon_k^+ + \epsilon_{k+}^- + \frac{2N}{N} \sum_p \bar{n}_p)$$

The use of $\langle\langle A^+, B^+ \rangle\rangle_\omega = - \langle\langle A, B \rangle\rangle_{-\omega}^*$ has been made in obtaining eqn (4.32). From this equation, it is clear that the line is of the Lorentzian type. However, it is known experimentally that at resonance the spin waves excited possess q ($\sim \frac{1}{\delta}$, δ being the skin depth) of the order of reciprocal skin depth. The width, therefore, should be given by

$$\Delta\omega = \frac{2 \chi_q(\omega)}{\hbar} \quad (4.33)$$

where $\chi_q(\omega)$ is given by eqn (4.26b) and has the significance of the linewidth. It is obvious from the expression for $\chi_q(\omega)$ that for finite non-vanishing linewidth the following energy relation should be satisfied

$$\omega - \epsilon_k^+ + \epsilon_{k-q}^- + \frac{2}{N} J_{s-d} \sum_p \bar{n}_p = 0 \quad (4.34)$$

The last term in eqn (4.34) is an additional term⁸⁷ and is absent from the corresponding equations of Vonsovskii et al⁴³.

The above energy conservation relation suggests that in the relaxation process a bunch of magnons are participating. Vonsovskii et al⁴³ have drawn attention to the fact that the relaxation process which results in spin flip of a conduction electron by emission or absorption of spin waves becomes ineffective for low energy spin waves due to the large exchange gap between bands of s electrons with different spin orientations and energy conservation law. However,

Heinrich et al⁴⁴ have recently suggested that non-stationary states of s spins make possible, in principle, a non-resonant absorption at a very low spin wave energy. It is quite possible that the thermal magnons denoted by eqn (4.27) might be associated with such non-stationary states of s-electrons.

It should be recalled here that the ferromagnetic 3d-electrons have been assumed to be localized and a Heisenberg type of exchange has been used to describe them. The 3d-electrons in ferromagnetic metals, specially Ni, are not localized but are known to be itinerant. Therefore, an itinerant electron model should be a realistic approach to the situation existing in these metals.

It will be interesting to mention here that recently Hubbard and Jain⁸⁸ have reported a calculation of the generalized spin susceptibility in a correlated narrow energy band model which is based on the narrow energy band theory given by Hubbard⁸⁹. According to this theory, a partly filled narrow energy band (s-band) is considered and it is assumed that the electron-electron Coulomb repulsion acts only between electrons of opposite spin on the same atom and has the magnitude I . They have discussed two special cases of their susceptibility function, namely, (1) weak interaction (band) limit, i.e., $I \ll \Delta$, where Δ is the band width and (2) strong interaction (atomic) limit, i.e., $I \gg \Delta$.

In the weak interaction limit their result reduces to

random phase result obtained by Izuyama et al.²⁰ and in the opposite limit a Curie law is obtained. Their theory is also not likely to explain linewidths in ferromagnetic metals.

It has already been pointed out that the spin wave energy is negligibly small compared to the energy required to flip the spin of the conduction electron. If the spin wave damping is to take place, as many as 10^3 or 10^4 spin waves would be required to participate in the relaxation process. This seems to be an impossibility. However, such processes, if they operate in these metals, should be reflected in spin wave resonance experiments. The superconducting ferromagnets may, perhaps, be the best materials to look for such processes

CHAPTER - V

CONCLUSION AND DISCUSSION

In the present work the interaction of the ferromagnetic 3d electrons in a ferromagnetic transition metal (Ni, Fe) interacting with the conduction s electrons by means of "s-d exchange" interaction has been considered to explain ferromagnetic relaxation. Using Green function formalism the expressions for the dynamic magnetic susceptibility and line width due to "s-d exchange" interaction have been obtained. It has been suggested that a bunch of magnons participate in the relaxation process resulting in the conduction electron spin flip. It should be noticed here that an exchange interaction of the Heisenberg type has been assumed between the magnetic and conduction electrons. This is not justified as the Heisenberg model assumes that the electrons are localized on the atoms at the lattice sites. The electrons in transition metals are not localized but are known to be itinerant (band) electrons. This is because of the fact that the 3d shell in these metals is on the outer side of the ion and, therefore, the electrons can move from one atom to the other forming a narrow band of states. A band model for magnetic electrons would, therefore, be a more realistic approach to the situation existing in ferromagnetic metals and the effect of conduction electrons should be properly taken into account.

The spin wave excitations in an itinerant electron model have been discussed. It is found that the spin wave energies in the long wavelength limit are proportional to k^2 (k being the wave vector of the spin wave) and thus have the same spectrum as the spin waves on the localized electron model. We thus have a probable explanation for the use of the localized electron model to a ferromagnetic transition metal

The renormalized spin wave and phonon energies resulting from the coupling of the spin waves with phonons have been obtained. It is found that the Bose-Einstein distributions for spin wave and phonon occupation numbers are "smeared out" because of magnon-phonon coupling

It should be mentioned here that the "s-d exchange" interaction can be used to investigate many other interesting properties such as conduction electron spin polarization, anomalous electrical resistance of transition metals and a very interesting phenomenon, i.e., the coexistence of ferromagnetism and superconductivity in some superconducting ferromagnets. It should be noticed that "s-d" or "s-f" (interaction between the conduction electrons and the 4f unfilled shell electrons) will act unfavourably to the simultaneous realization of the ferromagnetism and superconductivity. If the process of the spin wave damping results in spin flip of the conduction electrons then the "s-d exchange" will oppose the tendency of simultaneous occurrence of ferromagnetism and superconductivity, as the

absorption or emission of spin waves by, say, one of the electrons of the Cooper pair resulting in its spin flip will break the Cooper pairs and, therefore, will destroy superconductivity.

The theory discussed here is unable to explain linewidth and its temperature dependence in ferromagnetic metals. The reason for this is that it has not been possible to take the exact microscopic origin of the relaxation mechanism into consideration. In order to find the microscopic nature of the relaxation mechanism, more refined experiments on FMR in ferromagnetic transition metals must be performed, which can give some clue to the mechanisms in these metals.

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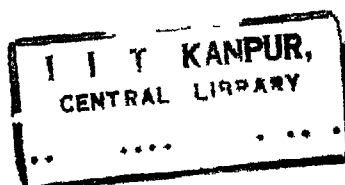
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